IUPAC Mole Project meeting minutes

25-28 July 2014

Kloster Kappel am Albis, Switzerland

EXECUTIVE SUMMARY

In the end of December of 2013, IUPAC approved a project proposal which aims to critically review the definitions for the quantity amount of substance and its SI unit, mole. At the present meeting, the Task Group focused mainly on the scientific and technical aspects as well as on the reasons of the current and the proposed definitions of the mole.

REMIT

This project aims to achieve internal IUPAC consensus on the definition of the mole. The outcome of this project is an IUPAC Technical Report which may or may not change the official IUPAC position on the mole which has been ratified by the IUPAC Council in 2011.

VENUE AND DELEGATES

IUPAC Project 2013-048-1-100 Task Group held its second meeting at the Kappel am Albis monastery near Zurich (Switzerland) on 25-28 July 2014. The following were present:

Task Group Members

Juergen Stohner (JS), *Zürich University of Applied Sciences (Switzerland); Chair of the Task Group, Chair of IUPAC Commission I.1, Secretary of IUPAC ICTNS, and Titular Member of IUPAC Division I*

Roberto Marquardt (RM), *University of Strasbourg (France); President of IUPAC Division I and Member of IUPAC Bureau*

Zoltan Mester (ZM), *National Research Council Canada; Secretary of IUPAC Division V*

Juris Meija (JM), *National Research Council Canada; Chair of IUPAC CIAAW and Titular Member of IUPAC ICTNS*

Marcy Towns (MT), *Purdue University (USA); Titular Member of IUPAC CCE*

Invited expert

Richard Davis (RD), *International Bureau of Weights and Measures (retired); Interim Executive Secretary of CCM (BIPM)*

Apologies

Ron Weir, *Royal Military College of Canada; Chair of IUPAC ICTNS*

Abbreviations used in this report

BIPM: International Bureau of Weights and Measures
CCE: IUPAC Committee on Chemistry Education
CCEM: CIPM Consultative Committee for Electricity and Magnetism
CCQM: CIPM Consultative Committee for Amount of Substance: Metrology in Chemistry
CCM: CIPM Consultative Committee for Mass and Related Quantities
CCU: CIPM Consultative Committee for Units
CGPM: General Conference on Weights and Measures
CIPM: International Committee for Weights and Measures
CIAAW: IUPAC Commission on Isotopic Abundances and Atomic Weights
CODATA: The Committee on Data for Science and Technology
ICTNS: IUPAC Interdivisional Committee on Terminology, Nomenclature and Symbols
IPK: International Prototype Kilogram
IUPAC: International Union of Pure and Applied Chemistry
IUPAP: International Union of Pure and Applied Physics
NAO: National Adhering Organization of IUPAC
SI: The International System of Units

– [Friday 25 July 2014] –

Arrival & informal meetings and discussions

– [Saturday 26 July 2014] –

1. OPENING OF THE MEETING AND THE ADOPTION OF THE AGENDA

Task Group Chairman (JS) opened the meeting at 10:20 and expressed his best wishes to the Task Group member Ron Weir (Chairman of ICTNS), who could not attend due to illness. Robert Wielgosz (BIPM) was invited but could not attend. Richard Davis (BIPM) attended on his behalf. The following agenda was adopted for the meeting:

1. Opening of the meeting (JS)
2. Approval of the Agenda (JS)
3. Introduction and NAO Questionnaire (JS)
4. Arguments for defining a fixed value of the Planck constant in the “new SI” (RD)
5. Questions and Discussion

6. The mole: its history, definition and realization in the “new SI” (RD)
7. Free discussions
8. Distribution of tasks
9. Contents of the Technical Report and initial evaluation of the literature
10. Free discussions
11. Closing of the meeting

2. INTRODUCTION AND QUESTIONNAIRE

JS gave an overview and explained the context of the project. Following an interdivisional meeting during the 47th IUPAC General Assembly in Istanbul, a project proposal was submitted to the IUPAC Secretariat regarding the critical evaluation of the proposed redefinition of the mole. The project proposal, entitled “A critical review of the proposed definitions of fundamental chemical quantities and their impact on chemical communities” was approved by IUPAC on 18 December 2013 with the objective to provide a Technical Report containing a critical review of the definitions for the quantity amount of substance and its unit, the mole, as well as the related unit of the quantity mass.

The Project will compile and critically evaluate the existing published work related to the definition of the quantity amount of substance, its unit, and the consequence of these definitions on the unit of the quantity mass, and vice versa. In addition, the project will perform a broad consultation on the said matters with the National Adhering Organizations of IUPAC.

Context

JS gave a brief historic overview in regards to the redefinition of the mole. On 9 July 2009, Prof. Ian Mills (the IUPAC Representative to, and President of, the CCU), sent a letter to IUPAC ICTNS Chair Prof. J. W. Lorimer informing him of CCU intentions to propose the redefinition of several SI base units including the mole. At the ICTNS meeting during the 45th IUPAC General Assembly in Glasgow, the implementation of the “New SI“ and its consequences were part of Agenda Item 8.2. Among the attendees were: John Dymond (Div. I), Jan Reedijk (Div. II), Amelia Rauter (Div. III), Richard Jones (Div. IV), Brynn Hibbert (Div. V), Peter Fedotov (Div. VI), Monica Nordberg (Div. VII), Joszef Nyitrai (Div. VIII), Ron Weir (ICTNS), Roberto Marquardt (ICTNS), Ales Fajgelj, Paul De Bievre, Ian Mills, Tyler Coplen, Anders Thor (ISO TC-12), and Franco Pavese. Documents have been circulated in advance of the meeting.

On 3 August 2009, Prof. Mills, presented definitions considered by CCU for the kilogram, ampere, kelvin, and mole to fix the numerical values of h, e, k, and N_A, respectively, and a new constant-explicit format for the formal definitions of the base units of the SI. This presentation was followed by a presentation of Paul de Bievre. Jack Lorimer, as Chair of ICTNS, put forward a motion which was adopted by vote following an extended discussion. This motion adopted the ICTNS resolution concerning the proposal by the CCU to redefine the mole and reads in its final form as follows:

> Given that:
>
> (a) definition of the mole in a way that is independent of mass is desirable;
> (b) the mole is often thought of by chemists as an Avogadro number of entities; and
> (c) the name of the ISQ base quantity "amount of substance" has been a source of much confusion, ICTNS recommends to the Bureau that:
>
> The recommendation of the CCU (Consultative Committee on Units) of the BIPM, that the mole be defined as follows:
>
> > "The mole, unit of amount of substance of a specified elementary entity, which may be an atom, molecule, ion, electron, any other particle or a specified group of such particles, is such that the Avogadro constant is equal to exactly $6.022\,141\,79\times10^{23}$ per mole.
> >
> > Thus we have the exact relation $N_A = 6.022\,141\,79\times10^{23}\ \text{mol}^{-1}$. The effect of this definition is that the mole is the amount of substance of a system that contains $6.022\,141\,79\times10^{23}$ specified elementary entities."
>
> be supported by the IUPAC, with the following suggestions:
>
> > 1. The greatest effort should be made to change the name of the ISQ base quantity "amount of substance" at the same time that a new definition of the mole is approved.
> >
> > 2. A note should accompany the new definition to explain that the molar mass of ^{12}C will be an experimental quantity, with a relative measurement uncertainty of about 1.4×10^{-9}.

(Note that the uncertainty 1.4×10^{-9} quoted by ICTNS was correct at the time, but an updated uncertainty is quoted on page 8 of the present report). On 26 August 2009, this was communicated to the IUPAC Bureau by the ICTNS Chairman Jack Lorimer. IUPAC Secretary General, Prof. Black, presented this resolution to the IUPAC Executive Committee at its 141st meeting of 3-4 October 2009 (Chile). A motion was made and seconded that the motion proposed by ICTNS be approved. The motion was approved unanimously by the IUPAC Executive Committee. This motion was communicated to BIPM as the official IUPAC position on that matter. The said resolution of the Executive Committee was ratified by the IUPAC Council during its 46th meeting of 3-4 August 2011 (Puerto Rico).

JS noted that whereas the first draft motion put forward by ICTNS Chair J. Lorimer stated that "The name preferred by IUPAC for the SI base unit is 'chemical amount'", the final motion adopted by vote states that "The greatest effort should be made to change the name of the ISQ base quantity 'amount of substance' at the same time that a new definition of the mole is approved."

(Added by JS after the meeting) JS also noted that in his letter from 9 July 2009 to J. Lorimer, I. Mills expressed his willingness to inform ICTNS at the Glasgow meeting about the proposal from CCU to redefine four base SI units among which

is the mole: “I would like to hear opinions from members of your committee. I certainly wish ICTNS to be aware of this proposal, and I would really like to come away with some expression of support for the proposal – or perhaps of opposition, if that should be the case. […] The CCU strongly supports making the change to fix the value of N_A. However some French chemists, championed by Yves Jeannin (a former president of IUPAC), are strongly opposed to making this change – they wish to retain the present definition of the mole.”

In September 2011, the Chairman of CIAAW sent a letter to the Secretary of CIPM representing that Commission’s own position which differs from the one officially communicated to BIPM by IUPAC. In particular, it “recommended” the redefinition of the kilogram as “the mass of one mole of ^{12}C atoms in their nuclear ground state multiplied by 1000/12” [as RD pointed out, the omitted specification of whether the atoms are bound or unbound has more significance than specifying their nuclear energy state] and a definition of the mole as “a number of specified entities equal to 6.022 14Xx10^{23} entities exactly”. This opinion expressed by CIAAW confused the BIPM which corresponded by asking the IUPAC President what is the official position of IUPAC given the CIAAW statement and the associated inconsistency. Consequently, the IUPAC Bureau reaffirmed explicitly the procedure followed by ICTNS. IUPAC Division V has since also voiced its concern over the official IUPAC position during the 2013 General Assembly. RM and ZM both attended the 19th meeting of CCQM in April 2013 from which RM felt that the definition of the mole in the “new SI” receives ambivalent support in CCQM and that it is perhaps not yet mature. These events have prompted the launch of the interdivisional IUPAC project soon after the 2013 IUPAC General Assembly to critically re-examine the proposed redefinition of the mole and the large amount of papers published on this topic.

At the ICTNS meeting during the 43rd IUPAC General Assembly in Beijing (2005), with the following attendees: J. Corish (Div. II), A.P. Rauter (Div. III), J. Kahovec (Div. IV), W. Kutner (Div. V), Y. Shiva (Div. VI), and A. McNaught (Div. VIII), Agenda Item 5.2 dealt with a polemic on the mole published in the *Journal of Chemical Education* during 2003 and 2004. This prompted the Editor J.W. Moore to note: “Henceforth I will entertain manuscripts from official groups whose purview is defining and naming units, where the manuscripts inform readers and allow for input to said groups, but I will not entertain manuscripts that initiate proposals for defining and naming units.” It was therefore considered that IUPAC should promote publication of an article (or a series) where the official IUPAC position regarding questions like the adoption of SI units and the redefinition of the mole or the kilogram should be explained.

JS also gave an overview of the procedures, bodies, and timelines involved in changing the definitions of SI units. To wit, the CGPM decides on changes to the SI at one of its meetings which usually occur every four years. CGPM is not a deliberative body and the resolutions voted by the CGPM are prepared by the CIPM following the advice of its Consultative Committees, CCU in particular. Given that CCU will meet in June 2015 to prepare the next edition of the SI Brochure, this Task Group must deliver its comments to IUPAC Bureau by March 2015 so that IUPAC may affirm or reformulate its position in a timely fashion. Therefore, it seems that a time horizon of March 2015 for having an established firm position of IUPAC with regard to this question is adequate for further decisions to be taken by CCU in 2015, CCQM in 2015 or 2016, CIPM in 2016 or 2017 and CGPM in 2018.

Questionnaire

In early June 2014, the Task Group initiated a survey to collect opinions and comments by the IUPAC National Adhering Organizations (NAOs). The questionnaire (see Appendix 1) was approved by the IUPAC Secretary General and it focuses on the current definition of the mole, the proposed new definition of the mole, the current definition of the quantity amount of substance, and the current name of the quantity amount of substance. Because of external time-constraints, IUPAC NAOs were asked to reply no later than 1 October 2014. As discussed in the letter to NAOs, all comments will be used towards formulating the IUPAC Technical Report. Those organizations who will reply will be sent an advance draft of the IUPAC Technical Report for further comment and input before submission to Pure and Applied Chemistry. After the presentation, all Task Group members filled the questionnaire and presented their personal views.

3. ARGUMENTS FOR DEFINING A FIXED VALUE OF THE PLANCK CONSTANT IN THE "NEW SI"

RD gave a talk entitled "Arguments for defining a fixed numerical value of the Planck constant, h: consequences for metrology in electricity, chemistry, and mass". The main points in the talk were: (1) the concept of "fixing" the numerical values of constants, (2) overview of modern electrical metrology, and (3) overview of the various possible choices for the revision of the SI base units kilogram, mole and ampere (and the derived units volt and ohm).

What does it mean to fix a constant of nature

Since 1983, the numerical value of the speed of light in vacuum, c, has been fixed (defined to have an exact value). Although the SI has fixed the numerical value of c, it says nothing about the properties of electromagnetic radiation. The numerical value does, however, say something important about the SI unit m/s. The SI now defines the magnitude of the unit m/s in terms of c, and a different numerical value would result in a different m/s but not a different c. In addition, the fixed value was not chosen to be, for example, 300 000 000, but 299 792 458, because historical continuity of the m/s (and the m) had to be maintained by the 1983 redefinition.

Currently, there are plenty of examples of fixed numerical values in the present SI or outside the SI. For example, the fixed numerical value of the hyperfine splitting frequency of the caesium-133 atom will continue to define the second, and the fixed numerical value of the mass of carbon-12 nuclide defines the atomic mass unit (the dalton) upon which the atomic-weight scale is based.

Modern electrical metrology

The electrical metrology community, represented by the CCEM, and the IUPAP have proposed to fix the numerical values of two constants that, at present, are determined experimentally: the elementary charge (e) and the Planck constant (h). These changes were approved in principle in 2011 by the CGPM. The reasons for such a proposal stem from the nature of modern electrical standards. The electrical potential difference (electric tension) is now measured most accurately via the Josephson effect (1973 Nobel Prize in physics) and electrical resistance is now measured most accurately by the quantum-Hall effect (1985 Nobel Prize in physics). In addition, the ampere can be realized from Ohm's law. Both of these quantum effects involve proportionality coefficients between the electrical quantities and the quantum numbers (and microwave frequency for voltage) – the Josephson constant, $K_J = 2e/h$, and the von Klitzing constant, $R_K = h/e^2$. Currently, the SI value of K_J is known with a relative standard uncertainty of 2200×10^{-11} whereas Josephson arrays can produce 10 V, with relative precision 5×10^{-11}. For these reasons, the electrical metrology community (CCEM) adopted conventional values for the Josephson constant ($K_{J\text{-}90}$) and von Klitzing constant ($R_{K\text{-}90}$) in 1990, based on CODATA-1989 SI values, but with the uncertainty set to zero. This led to a conventional volt, V_{90}, and a conventional ohm, Ω_{90}. After 20 more years of progress, the conventional values are offset from the currently recommended SI values (CODATA-2010), e.g. the SI volt now differs from V_{90} by -6200×10^{-11}. By adopting fixed numerical values of e and h, the uncertainties of K_J and R_K will vanish and the volt, ohm, and the ampere will be brought into the SI, with no need for conventional values. Electrical metrology will have to manage a noticeable, one-time-only change from conventional values to SI electric tension, resistance and current, but the long-term benefits are seen to outweigh the short-term inconvenience.

The Planck constant is the fundamental constant of quantum theory, just as the speed of light in vacuum is the fundamental constant of relativity theory. Many important effects are therefore described in terms of these constants so that fixed numerical values would be a benefit.

The kilogram will be redefined

The definition of the kilogram dates from 1889 and is not derived from a physical or atomic constant. This is no longer tenable in the 21st century because the magnitude of the SI unit of mass changes with the mass of IPK. There is general agreement that we must redefine the kilogram in terms of a constant because the means finally exist to do it and because that will provide the long-term stability of the kilogram. A consequence of fixing the numerical value of h (needed for K_J and R_K in electrical metrology) is that the kilogram becomes redefined. This is because the SI unit of the Planck constant is kg m^2 s^{-1}, and both the second and metre are already defined by physical constants.

Consequences and opportunities of fixed h

The Bohr-Sommerfeld model of the hydrogen atom links several fundamental constants. Today, all corrections to the model are incorporated in the Rydberg constant, R_∞:

$$2hcR_\infty = m_e(c\alpha)^2,$$

where m_e is the rest mass of electron and α is the fine structure constant. Relations for a particle X, where X may be an electron, an atom of ^{12}C etc., follow directly via $A_r(X) = m_a(X)/m_a(^{12}C)$:

$$h/m_a(X) = A_r(e)c\alpha^2/(2A_r(X)R_\infty) \text{ or}$$

$$N_A h/M(X) = A_r(e)c\alpha^2/(2A_r(X)R_\infty)$$

RD noted that this equation has been verified by the measurement of $h/m_a(^{87}Rb)$ in atomic recoil experiments. The relative uncertainty of the right-hand side of these equations is currently 7.0×10^{-10} for X = ^{12}C. The following conclusions are easily drawn:

(1) in the present SI, the numerical value of $M(^{12}C)$ = 12 g/mol is fixed so that the molar Planck constant N_Ah has uncertainty $u_r = 7.0\times10^{-10}$ (CODATA-2010)

(2) either the numerical value of h or m_u can be fixed, but not both. The uncertainty of the quantity that is not fixed is $u_r = 7.0\times10^{-10}$; too large for K_J.

(3) numerical values of N_A and h can be fixed (according to CGPM Resolution 1, 2011) and M_u and m_u will each have $u_r = 7.0\times10^{-10}$ when expressed in SI units.

(4) numerical values of M_u and h can be fixed (Jeannin suggestion) and N_A and m_u will each have $u_r = 7.0\times10^{-10}$ when expressed in SI units.

(5) numerical values of N_A and M_u can be fixed (which leads also to a fixed m_u) thus leading the kilogram to be redefined in terms of mass of carbon-12 atom and $u_r(h) = 7.0\times10^{-10}$ when expressed in SI units; too large for K_J.

Honouring the wish to change the definition of the kilogram, and honouring the needs of electrical metrology, one is left with two possible choices: the CGPM choice or the Jeannin choice. RD noted that the CCM, as a body, had expressed no preference for the chosen constant in the redefinition of the kilogram, as long as the new definition of the kilogram would allow the unit of mass, the kilogram, to be realized in practice to a relative standard uncertainty not greater than 2×10^{-8} by at least one method, and there is no significant difference between the kilogram as realized by either the watt balance or silicon x-ray crystal density methods. In short, the fixed Planck constant is required by the electrical metrology community and this choice also redefines the kilogram in a manner that satisfies CCM.

It was noted that the debate on the redefinition of the mole is not about a need for more accurate chemical measurements in SI units and this situation is markedly different from the fields of electrical metrology and mass metrology. The current debate about the redefinition of the mole thus focuses entirely on different questions, such as effective pedagogy. ZM initiated a discussion on the arguments for redefining the mole. It was noted that, once h is fixed, neither the electrical community nor the mass community has any further impact on the redefinition of the mole.

4. THE MOLE: ITS HISTORY, DEFINITION AND REALIZATION

On behalf of R. Wielgosz, RD gave an overview of the history of the mole. It was noted that the definition of the quantity for which the mole is the SI unit has not always been easy to describe in words. It is understood, however, that this quantity is a measure of an amount of matter which is proportional to the number of specified entities. A discussion arose as to whether a verbal definition of this quantity is needed given that many other quantities such as the mass or time are not verbally defined in the International System of Quantities. JM noted that we should if we can. Likewise, the name of the quantity has been inadequate and alternative proposals for the name have not led to popular use. A discussion arose regarding many proposed names and RM noted that "chemical amount" is not a well understood term. No conclusion was reached in this regard.

JM pointed out that since there is no technical need to redefine mole, one might ask what is gained by doing so. It is commonly viewed that a definition in terms of a fixed N_A leads to a conceptually simpler understanding of the mole, yet we give up the exact proportionality between the atomic weights and molar masses – something which, in turn, blurs the conceptual clarity just gained. RD suggested that it is important to distinguish between the (conceptual) definition of a unit and practical limits to its realization. A discussion also ensued regarding the utility of knowing the exact number of entities in a mole (CGPM-proposed definition of the mole) as opposed to knowing the exact mass of all those entities in a mole (current definition of the mole). JM commented that from a philosophical point-of-view, the current definition with its 'as many as' entities better reflects the nature of chemistry at-work as opposed to 'this many' entities in the proposed definition. After all, chemists are not concerned with the number of entities. Rather, they perform stoichiometric calculations and often involve equivalence tests (as in titrimetry or many other methods of analysis) in their work. On the contrary, RM felt that the exact number of entities in a mole is desirable feature which the current definition of the mole does not provide. ZM commented on the general lack of publications supporting the CGPM proposal regarding the redefinition of the mole authored by non-BIPM staff or by persons not directly involved in crafting the new proposals. ZM asked that a conscious effort be made to identify more such publications.

It became evident from RD presentation that fixing the numerical value of the elementary charge and that of the Planck constant, as well as having the *mise-en-pratique* of the definition of the kilogram based on a fixed numerical value of the Planck constant, are scenarios that are independent of the debate as to whether the mole shall continue to be defined as it is at present or whether it shall be redefined by fixing the numerical value of the Avogadro constant.

5. DISTRIBUTION OF TASKS

At the time of the meeting, the task group had 68 manuscripts at its disposal dating from 1961 to 2014. All manuscripts were assigned among the members for detailed inspection. It was agreed that all assigned manuscripts will be examined before the next meeting and a short internal report written for each publication. Meeting followed by informal discussions well into the night.

– [Sunday 27 July 2014] –

6. CONTENTS OF THE TECHNICAL REPORT AND EVALUATION OF THE LITERATURE

JS opened the meeting at 10:30 by distributing the table of contents of the Technical Report. It was agreed that the technical report will reflect the structure of the open letter sent to all IUPAC NAOs. The following outline was agreed:

1. Historical introduction (JM)
2. New SI which will outline the recent proposals to change the SI
3. Review of the literature with categories of published manuscripts
4. Critical evaluation of the literature
5. Questionnaire and the results
6. Synthesis

Lengthy discussions followed regarding the literature evaluation and the Task Group agreed on a template and approach to evaluate publications at hand. In particular, the publications will be categorized in one of the five categories outlined by RD (Agenda Item 3).

7. FREE DISCUSSIONS

Due to the free nature of most discussions, it was not feasible to record all issues raised during the meeting. However, several main points merit a note (in no particular order). ZM commented that the desire to uncouple the kilogram from the mole is subjective and does not merit the attention it has received. There are other examples of unit couplings in the SI (metre and second) and the reason to desire a mole definition independent from the kilogram, as suggested by CIPM, has no technical basis. MT noted that both the current and the new definition of the mole are difficult to teach and commented that our ability to understand the definition of the mole does not hamper our ability to do stoichiometry calculations. JS mentioned that the fact that one cannot teach something should not prompt us to change established concepts or definitions in the first place. RD commented that it is a common misconception that fixing *h* to define the kilogram means that realizing the kilogram is possible only via the watt balances. JM noted that the proposed definition of the mole (CGPM resolution) invokes the Avogadro constant and yet the current definition of the latter invokes the mole. This situation can be avoided by providing a better formulation for either the mole or the Avogadro constant. The Task Group agreed and RM sought to propose a more suitable and less entangled descriptions of the amount of substance and the mole. ZM also reflected on the inherent struggle between the access to the units which requires simple definitions, and our ability to perform high-precision measurements in these units which require sophisticated definitions.

Although the Task Group discussed pros and cons of the proposed new definition of the mole and some concluded that the proposed definition is not necessary from a technical point-of-view, not all arguments are reproduced in these minutes and therefore no conclusive statements should be drawn from these minutes before the Technical Report is completed.

8. CLOSING OF THE MEETING

JS closed the meeting in the evening and expressed his gratitude to RD for his efforts to explain the technical details behind many aspects of the proposed changes to the SI. The next meeting will take place in Ottawa (Canada) in January/February 2015.

Juris Meija

Secretary of the Task Group [Ottawa, October 14, 2014]

Corrected "CCEM" to "CCM" in Section 3 [November 28, 2014]