

February 24th, 2014

To Professor Ian Mills:

Thank you for your response to my Comments on Working Document CCU/13-09.3 and your efforts to enlighten me on the basics and nuances of the SI.

With respect to the former, you have clearly misinterpreted most of my points and invented things I did not say, and then berated me for the inconsistencies caused by your misinterpretations and inventions. You have evidently erroneously equated my (*dimensional*) "entity" ("ent") with your (*dimensionless*) "molecule" ("mcl"). My "entity" is entirely equivalent to Cesare Curti's "elementary entity" ("ee"), reference [5] in my Comments, and Morikawa & Newbold's "chemical amount for a single elementary entity, n_a , . . . which is the inverse of the Avogadro constant"—referenced below—the smallest possible amount of any substance and therefore the obvious choice for an atomic-scale unit for this (*dimensional*) quantity (not a "base" unit, and certainly not equal to the number 1). It is also a defining constant for relating the amount of a specified substance (or collection of specified entities), $n(X)$, to the corresponding number of entities, $N(X)$. The relationship is: $n(X) = N(X) \text{ ent}$ —i.e., the amount of a specified substance is an aggregate of $N(X)$ entities. Note that $\dim[n(X)] = \text{"AoS,"}$ $\dim[N(X)] = 1$, and $\dim(\text{ent}) = \text{"AoS."}$ The entity is also the defining constant for the macroscopic (SI base) unit, the mole: $\text{mol} = (\text{g/Da}) \text{ ent}$ —the mole is an Avogadro number of entities, where the Avogadro number is the gram-to-dalton mass-unit ratio, stemming from expressing substance mass in terms of grams rather than daltons, observing the mathematical identity: $\text{g} \equiv (\text{g/Da}) \text{ Da}$. This is exactly equivalent to the current definition, which can also be written explicitly in terms of the Avogadro constant, $N_A = 1 \text{ ent}^{-1}$ (one per entity): $\text{mol} = (\text{g/Da})(1/N_A)$. Your "molecule" is apparently some kind of dimensionless descriptive label, equivalent to 1, that can be multiplicatively inserted or extracted at will, reminiscent of the so-called "Factor-Label" method and the bizarre now-you-see-me-now-you-don't SI interpretation of the radian (where plane angle is supposed to be a "ratio of two lengths") and steradian (a square radian), this interpretation being the cause of widespread confusion about plane and solid angle.

I implore you to refrain from using the term "entity" and the symbol "ent" as equivalents of your "molecule" and "mcl," respectively. Below, I respond to your criticisms in detail.

Regarding your efforts to enlighten me about the SI, allow me to give you a little personal history. In the early 1970s, when I was a professor of Engineering Science at a branch of CUNY, the ASME (of which I was a member) and other engineering societies began an effort recommending a transition to the "metric system" within US technical education and industry. Our Division of Science and

Engineering quickly and seamlessly made the transition to "fully SI" instruction, including textbooks and laboratory equipment. As a consultant to the ASME, I was asked to edit some of their little brochures on "metric transition" in the areas of fluid mechanics, thermodynamics and heat transfer. Frankly, I was surprised by the number of technical blunders in these publications, mostly concerning conversion factors—such as confusing pound-mass, pound-force and slug, so that, for example, their published conversion factor for the universal gas constant was off by a factor of 32.174. They abandoned distribution of these poorly prepared brochures soon thereafter.

In 1985, when I came to The University of Akron, I was shocked to find that my new colleagues were still teaching mostly in US Customary units—the rationale being a perfect example of a vicious circle: (i) "In engineering colleges, we teach US Customary units because this is what our graduates need in industry." (ii) "In industry, we use US Customary units because this is what we were taught in college"! I single-handedly led an effort within the Faculty of Engineering to achieve fully SI instruction within all Departments by the mid-1990s.

About a dozen or so years ago, after some personal correspondence on various subtle nuances of SI terminology and symbols, Bruce Barrow invited me to join the IEEE SCC14 subcommittee on Quantities, Units and Letter Symbols. I helped with detailed technical editing of the IEEE/ASTM SI 10 brochure. In 2005, Barry Taylor, also a member of SCC14, alerted the committee chairman to the recent publication of "Redefinition of the kilogram: a decision whose time has come," MMQW(05) published in *Metrologia* in March of that year, and this was distributed to committee members. In that paper you and your co-authors considered two possibilities for redefining the kilogram:

- (1) fix N_A (retaining $M(^{12}\text{C}) = 12 \text{ g mol}^{-1}$, taken for granted)—thereby defining the kilogram as an exact multiple of the carbon-12 atomic mass (with the carbon-12-based dalton exactly related to this kilogram—although not mentioned explicitly), in which case, the Planck constant would (continue to) be determined by experiment; or
- (2) fix h (retaining $M(^{12}\text{C}) = 12 \text{ g mol}^{-1}$, taken for granted), in which case, the Avogadro constant would (continue to) be determined by experiment (the carbon-12-based dalton remains inexactly known in terms of this kilogram).

Since $h N_A = \text{const } M(^{12}\text{C})$, where const is inexactly known, it was immediately clear that there was a third possibility, not addressed in your paper:

- (3) fix both h and N_A —necessarily relaxing the exactness constraint on $M(^{12}\text{C})$, which would then have to be determined by experiment. However, in order to avoid creating an incompatibility between the kilogram, dalton and mole, fundamentally related by $\text{mol} = [(0.001 \text{ kg})/\text{Da}](1/N_A)$ —i.e., the mole is an Avogadro number of entities (or "reciprocal Avogadro constants")—the dalton would necessarily have to be redefined exactly in terms of the (fixed- h) kilogram,

so that $D_a = (1/1000N^*)$ kg, exactly, where N^* is a fixed specified number, in which case: $\text{mol} = N^*$ ent, an exact Avogadro number of entities.

In an effort to point this out to you and your colleagues, on March 21st, 2005, I sent a letter to you, with cc's to your co-authors, Peter Mohr, Terry Quinn, Barry Taylor and Edwin Williams, in addition to Jim Frysinger, Stan Jakuba, Bruce Barrow and Richard Davis, outlining in detail this third possibility—letter attached. In the letter, I also introduced my definition of ". . . an 'atomic-level' unit for amount of substance paralleling dalton, the atomic-level unit for mass. This unit would (obviously) be called entity (symbol ent). Thus, one entity (1 ent) is the amount of substance consisting of exactly one (specified) elementary entity." I also explained the use of "amount-specific" as the appropriate and self-explanatory generic adjective referring to a specific-intensive quantity in the form of the quotient (any extensive quantity)/(amount of substance), thereby replacing the Green-Book-rule-violating term "molar." I also recommended fixing the Boltzmann constant—" . . . why not immediately?"—and the elementary charge, both of which you had mentioned in MMQW(05).

Since I had no response from you or any of your colleagues, I decided to send a Letter to the Editor of *Metrologia* outlining my suggestion of the possibility of fixing both h and N_A , while "freeing" $M(^{12}\text{C})$ —and redefining D_a compatibly as an exact submultiple of kg: "Note on invariant redefinitions of SI base units for both mass and amount of substance." This was received June 2nd, 2005, but not published until December 22nd, 2005. I assume that you and your "New SI" colleagues would have been interested in and thoroughly familiar with my LTE when it appeared.

In the interim, Peter Mohr and Barry Taylor had produced "On defining the mole so as to fix the value of the Avogadro constant N_A when the kilogram is defined so as to fix the value of the Planck constant h ," submitted to the 17th CCU meeting, 29 June–1 July, 2005, Working Document CCU/05-29. Although Mohr and Taylor would have earlier received my March 21st letter, in which the fixed- h /fixed- N_A /exact D_a combination was spelled out, I have to assume that their proposal for the fixed- h /fixed- N_A (but incompatibly retained carbon-12-based dalton) combination was purely coincidental. Then in January 2006, you and your co-authors submitted to *Metrologia* the paper that has become the basis for the New SI, "Redefinition of the kilogram, ampere, kelvin and mole: a proposed approach to implementing CIPM recommendation 1 (CI-2005)," published in April, 2006, MMQW(06). Since you did not cite my March 21st letter (as "personal communication"—I know *Metrologia* doesn't like such citations) or my LTE (which had already been published the previous December), I have to assume that you and your colleagues (particularly Mohr & Taylor) came up with the fixed- h /fixed- N_A idea independently.

The main difference between your MMQW(06) proposal and my suggestion concerns the incompatibility of retaining the carbon-12-based dalton (which you evidently took, and still take, for granted), thereby requiring the introduction of a "correction factor," originally known as " $(1 + \kappa)$," as described by Mohr & Taylor. Since this proved to be confusing to many people, you and your colleagues (including Taylor and Milton) came up with the idea of imbedding the inexactly known correction factor within a "modified molar mass constant," so that $(1 + \kappa) = M_u/(1 \text{ g mol}^{-1}) = M(^{12}\text{C})/(12 \text{ g mol}^{-1})$. And this is supposed to be less confusing?

I have written a number of other papers criticizing this incompatibility—and offering compatible solutions—published in *Metrologia* and *ACQUAL*. I assume that you and your colleagues would be thoroughly familiar with these publications (and those of other authors commenting on the New SI), particularly Reference [2] in my Comments, as this paper is listed under "Discussion in the scientific literature" on the BIPM-New SI website: "Comments on recent proposals for redefining the mole and kilogram," *Metrologia*, April, 2010. As in previous papers of mine dealing with the entity as an atomic-scale unit of amount of (any) substance (or chemical amount), this paper also explains my definition of one entity as the smallest possible amount of (any) substance, so that $n(X) = N(X) \text{ ent}$ —the amount of a specified substance is an aggregate of $N(X)$ entities. When $N(X) = \text{g/Da}$, the Avogadro number, this defines the macroscopic (SI base) unit: $\text{mol} = (\text{g/Da}) \text{ ent}$: the mole is an Avogadro number of entities—a direct definition preferred by IUPAC and CIAAW—"as it is often thought of by chemists." The "explicit-unit" form of the New SI definition is: $\text{mol} = N^*(1/N_A)$ —i.e., the mole is an exact fixed number of "reciprocal Avogadro constants." Since $N_A = N(X)/n(X) = N(X)/[N(X) \text{ ent}] = 1 \text{ ent}^{-1}$ (one per entity), then $1/N_A = 1 \text{ ent}$, so the New SI mole is an exact fixed number of entities: $\text{mol} = N^* \text{ ent}$. But, with the fixed- h kilogram, N^* is not equal to the Avogadro number, g/Da , that arises (entirely) from the substance mass expressed in grams rather than daltons—thus creating the incompatibility and the need for a " $(1 + \kappa)$ -type" correction factor, no matter how it is disguised. This is not the way to design a 21st-century units system!

Since N_A is a defining constant, so too must $1/N_A = \text{ent}$ be a defining constant.* N_A has the incomprehensible dimension of "reciprocal amount of substance," whereas ent has the appropriate dimension of amount of substance. Doesn't it therefore make much more sense to work directly with a defining constant for amount of substance rather than a defining constant for "reciprocal amount of substance"? Formally, the results are the same of course; but an aggregate of a number of entities is far easier to understand than an aggregate of a number of "reciprocal Avogadro constants."

Let me reiterate: one entity (the reciprocal of the Avogadro constant)* is the smallest possible amount of (any) substance—a "natural" and easily understood defining constant, with the physical dimension of amount of substance (or chemical amount). It is therefore the appropriate atomic-scale unit for the amount of (any) substance; it is not the number 1, nor can it be replaced by 1—just as $1/N_A$ (= 1 ent) is not 1, nor can it be replaced by 1. The SI base unit, the mole, is then defined quite transparently as: mol = (g/Da) ent—an Avogadro number of entities, "as it is often thought of by chemists." Since I assume that you would be quite familiar with at least my paper referenced on the BIPM-New SI website, I find your responses to my "Comments" quite puzzling, to say the least.

Thank you for sending along sections of the Draft revised 9th edition of the BIPM Brochure. Actually, I already had a copy and have sent suggested "corrections" to Dr Wielgosz, who sent these on to Professor Ullrich. I will forward these to you separately for your reference. Lastly, you can be assured that whenever I am working on SI-related matters, I keep my copy of the Green Book (3rd edition) on my desk or within easy reach, along with relevant printed sections of the BIPM Brochure.

Yours sincerely,

Benny Leonard

Dr B P Leonard
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Reference:

Morikawa T and Newbold B T (2004) 'Amount of substance' and a single elementary entity. *Chemistry (Bulgarian Journal of Chemical Education)* 13:431–435

*In his *Phil Trans Roy Soc* article, "A new definition for the mole based on the Avogadro constant: a journey from physics to chemistry," Martin Milton states that with "the new definition of the mole . . . the amount of substance corresponding to one entity would be $\{N_A\}^{-1}$ exactly"—where the curly brackets indicate the numerical value of the Avogadro constant (understood to be expressed in reciprocal moles). The inclusion of the curly brackets is presumably a typographical error. Since $n(X) = N(X)/N_A$, the correct expression is: $n(1 \text{ entity}) = 1/N_A$, not $1/\{N_A\}$.

Commentary on Professor Mills's response to my Comments on Working Document CCU/13-09.3

- **The name “chemical amount”**

We agree that this is a better name than “amount of substance.” The actual concept is “amount”: amount of water, $n(\text{H}_2\text{O})$; amount of monatomic oxygen, $n(\text{O})$; amount of a collection of electrons, $n(\text{e})$; &tc. But, just as there are different kinds of “currents,” there are also different kinds of “amounts” (not many). Adding the adjectives “electric” and “chemical” for the respective formal names of these physical quantities is just common sense—with the understanding that, in practice, the adjectives can usually be omitted when there is no chance of confusion with other kinds of currents or amounts.

- **The entity and the mole**

Professor Mills says that I “wish to introduce ‘entity’ as the base unit for AoS, and somehow treat the mole as a sort of derived unit equal to $6.022 \dots \times 10^{23}$ entities.”

This statement is not correct! I said no such thing.

The entity, being the smallest amount of any substance, is the appropriate *atomic-scale* unit for chemical amount, paralleling the dalton as the appropriate atomic-scale unit for mass. Neither one is a *base* unit. The entity takes the place of the Avogadro constant as a defining constant—for chemical amount rather than for “reciprocal chemical amount,” respectively. Thus we have the following fundamental relationship between $n(\text{X})$ and $N(\text{X})$:

$$n(\text{X}) = N(\text{X}) \text{ ent} \tag{1}$$

rather than the current SI version:

$$n(\text{X}) = N(\text{X})(1/N_{\text{A}}) \tag{2}$$

And the definition of the mole:

$$\text{mol} = \mathcal{N}_{\text{Avo}} \text{ent} = (\text{g/Da}) \text{ent} \quad (3)$$

rather than the current (explicit-unit) definition:

$$\text{mol} = (\text{g/Da})(1/N_{\text{A}}) = 0.012m(\mathcal{K})/M(^{12}\text{C}) \quad (4)$$

All of these equations are dimensionally consistent.

- **Difference between N and n**

Professor Mills says that I “wish to have only one quantity that embraces N and n as being dimensionally the same.”

This statement is certainly not correct.

As can be seen from equations (1) and (2):

$$\dim(\text{ent}) = \dim(1/N_{\text{A}}) = \mathbf{C} \quad (\text{chemical amount}) \quad (5)$$

(or \mathbf{N} , according to the current SI). So, since $\dim[N(X)] = \mathbf{1}$, we have $\dim[n(X)] = \mathbf{C}$.

- **Amount-specific quantities**

Consider two *extensive* quantities, Q_1 and Q_2 . We can form what is called a *specific-intensive* quantity from the quotient of the two extensive quantities, say:

$$q_1 = Q_1/Q_2 \quad (6)$$

In general, q_1 is called the “ Q_2 -specific Q_1 ” (or the “denominator-specific numerator”). Such specific-intensive quantities are common in the fluid and thermal sciences, where

many different variables are involved. For example, if $Q_1 = m$ (mass) and $Q_2 = V$ (volume), we have:

$$\rho = m/V \quad (7)$$

the “volume-specific mass.” This has a special name: “mass density,” usually contracted to “density” if there is no chance of confusion with other kinds of density. As another example, the “mass-specific volume” is:

$$v = V/m \quad (8)$$

This is usually called “specific volume,” following a common custom: if mass is involved the adjective “mass” can be omitted—if there is no chance of confusion. Now consider:

$$v^* = V/n \quad (9)$$

where n is amount of substance or chemical amount. The full name would be “amount-of-substance-specific volume” or “chemical-amount-specific volume”—obviously a mouthful in either case. Since (as argued above) the actual quantity involved in the denominator is “amount,” the name of v^* is “amount-specific volume.” An amount-specific quantity is any extensive quantity that has been divided by n . In particular, $m(X)/n(X)$ is the amount-specific mass, $M(X)$. [It is certainly not “AoS/mass”! Rather, $n(X)/m(X)$ is “mass-specific amount” or “specific amount.”]

- **Textbooks of physical chemistry**

Professor Mills cites Peter Atkins’s well-known books as appropriate physical chemistry textbooks. In the Atkins & de Paula textbook *Physical Chemistry* (8th edition), page 156, they write: Molar masses of macromolecules are often reported in daltons (Da), with

$$1 \text{ Da} = 1 \text{ g mol}^{-1} \quad (10)$$

Inverting this means that Atkins & de Paula effectively define the mole as:

$$\text{mol} = (\text{g/Da}) = 6.022 \dots \times 10^{23} \quad (\text{Dimension: } \mathbf{1}) \quad (11)$$

—i.e., the Avogadro *number*, not an Avogadro number of *entities* or an Avogadro number of *reciprocal Avogadro constants* as in the current SI definition. If we still consider N_A to be the constant defining the relationship between $n(X)$ and $N(X)$ as:

$$n(X) = N(X)/N_A \quad (12)$$

with the mole defined by $n(X) = 1 \text{ mol}$ when $N(X) = \text{g/Da}$, then:

$$\text{mol} = (\text{g/Da})/N_A \quad (13)$$

as usual. Inverting this to give the Avogadro constant:

$$N_A = (\text{g/Da})/\text{mol} = (\text{g/Da})/(\text{g/Da}) \equiv 1 \quad (14)$$

which implies:

$$n(X) = N(X) = N(X)/(\text{g/Da}) \text{ mol} = N_r(X) \text{ mol} \quad (15)$$

where $N_r(X)$ is the “relative number of entities” as proposed by Peter Nelson. For amount-specific mass, we have:

$$M(X) = m(X)/n(X) = N(X)m_{\text{av}}(X)/N(X) = m_{\text{av}}(X) \quad (16)$$

—i.e., the amount-specific mass of a specified substance is equal to the (sample-average) entity mass of that substance. This is the “mole-as-a-number” concept, as explained in detail in the Appendix of reference [1] of my Comments and in the latter section of reference [4].

Note that if the entity were to be adopted as the atomic-scale unit of chemical amount, Atkins & de Paula could write, “Amount-specific masses of macromolecules are often reported in daltons per entity, Da ent^{-1} , where $\text{Da ent}^{-1} = \text{g mol}^{-1} = \text{kg kmol}^{-1}$, exactly.” Inverting this for the mole gives: $\text{mol} = (\text{g/Da}) \text{ent}$, an Avogadro number of entities. All of these relationships are dimensionally consistent. [In the 9th edition of *Physical Chemistry*, Atkins & de Paula stress that *molecular* (not molar) masses are measured in daltons, although they are commonly called “molar” masses.]

- **Appendix**

In the second paragraph of Professor Mills’s Appendix, he says that “the Avogadro constant . . . has the dimension of reciprocal mole in SI.” I assume he means “has the dimension of *reciprocal amount of substance*” or “has the *unit* of reciprocal mole (in SI base units).” [A *dimension* cannot be expressed in *units*.] As is (hopefully, by now) obvious from the above discussion, $n(X)$ and $N(X)$ are **not** “dimensionally the same.” Also, \mathcal{R} and k_B are different quantities with different dimensions and different units. Similarly for F and e .

- **Recommended notation for quantities**

On this sheet Professor Mills has listed several items in red involving “ent.” These should be deleted as they are ***absolutely false in all cases***—using my definition of entity (ent). In the footnote Professor Mills claims that the “unit entity, symbol ent, . . . is the same as the unit molecule, symbol mcl” ***This is absolutely false (using my definition of entity)***. He says that “Entity is perhaps a better name . . . In the SI this unit is equal to 1.” ***Again, this is absolutely false (using my definition of entity)***.

- **Plea to Professor Mills**

Please do not use the name “entity” as an alternative or substitute for your “molecule” or the symbol “ent” as an alternative or substitute for your “mcl.” This would undermine my carefully constructed and dimensionally consistent concepts and create untold further confusion!