

A view about the short histories of the mole and Avogadro's number

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Abstract The mole and Avogadro's number are two important concepts of science that provide a link between the properties of individual atoms or molecules and the properties of bulk matter. It is clear that an early theorist of the idea of these two concepts was Avogadro. However, the research literature shows that there is a controversy about the subjects of when and by whom the mole concept was first introduced into science and when and by whom Avogadro's number was first calculated. Based on this point, the following five matters are taken into consideration in this paper. First, in order to base the subject matter on a strong ground, the historical development of understanding the particulate nature of matter is presented. Second, in 1811, Amedeo Avogadro built the theoretical foundations of the mole concept and the number $6.022 \times 10^{23} \text{ mol}^{-1}$. Third, in 1865, Johann Josef Loschmidt first estimated the number of molecules in a cubic centimetre of a gas under normal conditions as 1.83×10^{18} . Fourth, in 1881, August Horstmann first introduced the concept of gram-molecular weight in the sense of today's mole concept into chemistry and, in 1900, Wilhelm Ostwald first used the term mole instead of the term 'gram-molecular weight'. Lastly, in 1889, Károly Than first determined the gram-molecular volume of gases under normal conditions as $22,330 \text{ cm}^3$. Accordingly, the first value for Avogadro's number in science history should be 4.09×10^{22} molecules/gram-molecular weight, which is calculated by multiplying Loschmidt's 1.83×10^{18} molecules/ cm^3 by Than's $22,330 \text{ cm}^3/\text{gram-molecular weight}$. Hence, Avogadro is the originator of the ideas of the mole and the number $6.022 \times 10^{23} \text{ mol}^{-1}$, Horstmann first introduced the mole concept into science/chemistry, and Loschmidt and Than are the scientists who first calculated Avogadro's number. However, in the science research literature, it is widely expressed that the mole concept was first introduced into chemistry by Ostwald in 1900 and that Avogadro's number was first calculated by Jean Baptiste Perrin in 1908. As a result, in this study, it is particularly emphasised that Horstmann first introduced the mole concept into science/chemistry and the first value of Avogadro's number in the history of science was 4.09×10^{22} molecules/gram-molecular weight and Loschmidt and Than together first calculated this number.

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Introduction

The International System of Units (SI) is based on the seven basic units. One of them is the mole. The mole is the unit of the amount of substance, which is the number of particles in a given mass. It was introduced into the SI unit system by the 14th General Conference on Weights and Measures (CGPM) in 1971 and is defined as the amount of substance in a system that contains as many elementary entities as there are atoms in 0.012 kg of carbon-12 (Lide 1992–1993, p. 1.14.). The number of atoms in exactly 0.012 kg of carbon-12 is called Avogadro's number (Hampel and Hawley 1982, p. 29; Pauling 1988, p. 96). The profound importance of Avogadro's number is that it provides a link between the properties of individual atoms or molecules and the properties of bulk matter.

There are many methods for the determination of Avogadro's number given in the research literature (Sarikaya 2004). The most modern method available today calculates Avogadro's number from the density of a crystal, the relative atomic mass, and the unit cell length determined by X-ray methods. Very accurate values of these quantities for silicon have been measured at the National Institute for Standards and Technology (NIST). Today's accepted experimental value of $6.022\,141\,29\,(27) \times 10^{23}$ atoms per mole is the best average for measurements using the best methods available (NIST 2011).

The atomic weight scale and the mole are two concepts which are related to each other and the order of precedence is the atomic weight scale. The mole is built on the atomic weight scale. Today, we know that the definitions of both the atomic mass unit and the mole are based on the carbon-12 isotope. The mole and Avogadro's number are also hierarchically related to each other and Avogadro's number is a result of the mole concept (Sarikaya 2004). Therefore, the mole concept was introduced to chemistry earlier than Avogadro's number. The mole concept was used for the first time by the German chemist August Horstmann (1842–1929) by the term 'gram-molecular weight' in 1881 (Cerruti 1994; Turco and Cerruti 2002). The German chemist Wilhelm Ostwald (1853–1932) used the term 'mole' instead of the term 'gram-molecular weight' in 1900 (Gorin 1994). Ostwald defined the mole as 'that amount of any gas that occupies a volume of 22,414 mL in normal conditions is called one mole' (Gorin 1994). The French physicist Jean Baptiste Perrin (1870–1942) introduced the expression 'Avogadro's number' into chemistry in 1908 (Becker 2001). Like Ostwald, Perrin also based the definition of the mole on the amount of substance in the volume 22.414 L of gases and he defined Avogadro's number as the number of molecules in one gram-molecular volume (namely 22.414 L) of a gas under normal conditions. In an effort to estimate the number of gas molecules in unit volume, in 1865, the Austrian chemist Johann Josef Loschmidt (1821–1895) calculated the number of molecules in a cubic centimetre of a gas under normal conditions as 1.83×10^{18} (Hawthorne 1970). After Ostwald's definition of the mole, and Perrin's definition of Avogadro's number, the early value of Avogadro's number was calculated by multiplying Loschmidt's 1.83×10^{18} molecules/cm³ by the value 22,414 cm³/gram-molecular weight. In order to estimate the early value of Avogadro's number, it is clear that the number 22.414 L mol⁻¹ has a great importance. However, we do not have definite knowledge about the facts of when and by whom the number 22.414 L mol⁻¹ was first determined. This value was measured by the Italian chemist Stanislao Cannizzaro (1826–1910) according to Wisniak

(2000), and by the Italian Chemist Amedeo Avogadro (1776–1856) according to Gladney (1999). These different expressions were even enough to show the fact that neither Avogadro nor Cannizzaro calculated the number 22.4 L mol^{-1} . Moreover, the concepts of the gram-molecular weight and gram-molecular volume, and the number 22.4 L mol^{-1} were implied in neither Avogadro's (1811) nor Cannizzaro's (1858) essays translated in the Alembic Club. However, literature research showed that the Hungarian chemist Károly Than (1834–1908) first determined the molar volume of gases as 22.33 L in 1889 (Than 1889). Accordingly, the first value for Avogadro's number in science history should be 4.09×10^{22} molecules/gram-molecular weight, which was calculated by multiplying Loschmidt's 1.83×10^{18} molecules/cm³ by Than's $22,330 \text{ cm}^3/\text{gram-molecular weight}$, ($22,330 \text{ cm}^3/\text{gram-molecular weight}$) ($1.83 \times 10^{18} \text{ molecules/cm}^3$) = 4.09×10^{22} molecules/gram-molecular weight. Hence, the scientists who first calculated Avogadro's number should be Loschmidt and Than.

One of the aims of science education is to help students in developing their feelings of respect and admiration about scientists. In this context, another aim is to guide students in becoming aware of the difficulties which scientists experienced. In addition, researchers have always indicated the importance of the history, nature and philosophy of science to learn science concepts (Adúriz-Bravo 2004; Erduran et al. 2007; Freire and Tenório 2001; Giunta 2001; Harrison 2002; Lin et al. 2002; Matthews 1989, 1992, 1994, 2007; Quílez 2004; Sarikaya 2007; Seroglou and Koumaras 2001; Watson 2007). However, many students maintain only a minimal interest in history and philosophy of science (Erduran 2005; Brush 1974). As is known, the first half of the nineteenth century has a very important place in the history of science. At that time, each of Dalton's, Gay-Lussac's and Avogadro's studies has a quality to be a milestone for the development of atomic theory. However, their studies are not clear for many people in science. When the researcher asked his students in general chemistry courses, and the chemistry teachers in the in-service training programmes, about why the number $6.022 \times 10^{23} \text{ mol}^{-1}$ is called Avogadro's number, almost all of them responded that it was due to the fact that the first scientist calculating this number was Avogadro. It is also stated in the same way in a Turkish science textbook (Aleaddinoglu et al. 1995, p. 123) for secondary students. Giunta (2011a) needs to emphasize the fact that Avogadro did not discover or determine or propose Avogadro's number because her students also had the same kind of difficulties. Although Becker (2001) indicates the importance of the number 22.4 L mol^{-1} in his article to calculate the value of Avogadro's number, he does not mention Than's name among the scientists who determine Avogadro's number. These facts show that people at all levels of chemistry/science are confused about the origins of the mole and the number $6.022 \times 10^{23} \text{ mol}^{-1}$. The definitions of the mole and Avogadro's number are based on the number $22.414 \text{ L mol}^{-1}$, but the facts of when and by whom this number was defined are overlooked. However, this number plays a key role in calculating Avogadro's number for the first time.

In this study, five things are pointed out. First, the historical development of understanding the particulate nature of matter was presented. Second, in 1811, Avogadro built the theoretical foundations of the number $6.022 \times 10^{23} \text{ mol}^{-1}$. Third, in 1865, Loschmidt first estimated the number of molecules in a cubic centimetre of a gas under normal conditions as 1.83×10^{18} (Hawthorne 1970). Fourth, in 1881, Horstmann introduced the concept of gram-molecular weight in the sense of today's mole concept into chemistry (Cerruti 1994; Turco and Cerruti 2002). Lastly, in 1889, Than first determined the gram-molecular volume of gases under normal conditions as $22,330 \text{ cm}^3$ (Than 1889). Accordingly, the first value for Avogadro's number in the history of science should be 4.09×10^{22} molecules/gram-molecular weight, which was calculated by multiplying

Loschmidt's 1.83×10^{18} molecules/cm³ by Than's 22,330 cm³/gram-molecular weight. Hence, Avogadro was the originator of the ideas of the mole and the number 6.022×10^{23} mol⁻¹, and Loschmidt and Than were the scientists who first calculated the number 6.022×10^{23} mol⁻¹. Avogadro and Loschmidt have been well-known figures in the science world. However, we never knew Than's role in determining Avogadro's number. The researcher suggests that the fact that Than is one of two scientists who first calculate the number 6.022×10^{23} mol⁻¹ should be acknowledged by the science world and that the name 'Károly Than' should be mentioned in textbooks in order to show our gratitude.

Background history

Because Avogadro's number is related fundamentally to atoms and molecules, perhaps it will be much more appropriate to begin with a brief discussion of the historical development of atomic theory. The first people who thought seriously about the nature of matter were the ancient Greek philosophers, about 2,500 years ago. In the fifth and fourth centuries BC, Leucippus (480–420 BC) and Democritus (460–370 BC) proposed that matter was made up of things called atoms and could not be endlessly divided into smaller particles (Matthews 1994; Partington 1960; Tsaparlis 2001).

The Greek atomism lacked the essential features of a scientific theory. It was not based on planned experimentation. Since it was a construct of conjecture, it could also be demolished by conjecture. The Greek theory dominated scientific thought for centuries, and in this background, atomic theory was abandoned for at least 2,300 years. Finally, it was reconsidered by the British chemist Robert Boyle (1627–1691) in the middle of the seventeenth century (Mahan 1972). Boyle was the first experimenter who accepted the existence of atoms in his book 'The Sceptical Chemist.' He defined the concepts of element and compound and believed in the atomic theory, which he used to explain chemical changes and gas pressure (Matthews 1994; Partington 1960, pp. 65–152). Towards the end of the seventeenth century, the British physicist Isaac Newton (1643–1727) gave place to the concept of atoms in his books 'Principia' and 'Opticks.' He also believed that elastic fluids (gases) are composed of small mutually repellent particles (Garrett 1968, pp. 68–72; Partington 1960, pp. 153–179). In the latter part of the eighteenth century, the French chemist Antoine Laurent Lavoisier (1743–1794) introduced quantitative measurements in chemistry. He found that there is no net change in weight during a chemical reaction (Partington 1960, pp. 122–152). Today, this principle is known as 'the law of conservation of mass.' At the beginning of the nineteenth century, the French chemist Joseph Louis Proust (1754–1826) found that the weight proportions of the elements in a pure compound are fixed (Partington 1960, pp. 153–179). Today, this finding is known as 'the law of definite composition.'

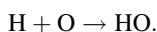
Incorporating the atomic ideas of the ancient Greeks, Boyle's and Newton's concepts of element and compound, Lavoisier's law of conservation of mass, Proust's law of definite composition with the law of multiple proportions and some other new ideas of his own, the British chemist John Dalton (1766–1844) developed an atomic theory, which is a landmark in the history of chemistry, between the years of 1803 and 1808. When developing his atomic theory, Dalton was concerned about the physical properties of gases (Garrett 1968, pp. 68–72). In an effort to explain diffusion of gases, he had considered Newton's hypothesis that gases are composed of small mutually repellent particles. In his work, Dalton got the idea that molecules of different gases have different volumes. In order to account for the different solubilities, he assumed that the bulk of gas dissolved depends

upon the weight and the number of particles. These observations were the original stimuli for his atomic theory. He formulated his theory into the following three basic principles (Partington 1960, pp. 153–179):

1. The chemical elements are composed of very minute indivisible particles of matter, called atoms, which preserve their individuality in all chemical changes.
2. All the atoms of the same element are identical in all respects, particularly in weight. Different elements have atoms differing in weight. Each element is characterized by the weight of its atom.
3. Chemical combination occurs by the union of the atoms of the elements in simple numerical ratios.

In this point, Dalton considered the problem of determining both atomic weights and molecular formulas. It is clear that if one of them were known, the other could be determined. In 1805, when hydrogen was burned in oxygen to form water, it became apparent that one gram of hydrogen combines with eight grams of oxygen. However, the weight ratio could not give an idea about the molecular formula of water since the relative weights of the hydrogen atom and the oxygen atom were not known. To make any progress, some assumptions were necessary. Dalton tackled this problem with an approach frequently employed in scientific investigation: if there is no information to the contrary, the simplest possible assumption is made and its consequences are pursued. He suggested the ‘Rule of Greatest Simplicity’: when two elements A and B form a compound, it is a binary AB; if more than one compound is formed, the first is a binary AB, and the next is a ternary A₂B or B₂A. Dalton, who thought that atoms of the same element repel each other, regarded a binary compound as the most stable one (Partington 1960, pp. 153–179). Based on the Rule of Greatest Simplicity, he assumed that the molecular formula of water was HO since hydrogen peroxide had not yet been discovered (Wernimont et al. 1999).

Dalton was able to develop an atomic weight scale which eventually proved to be in error because he assumed that particles of the common gaseous elements are composed of single atoms. For the reaction between hydrogen and oxygen, he wrote



Dalton assigned a weight of 1 unit to the hydrogen atom and calculated the relative weights of the atoms of the other known elements (Griffith 2010). He was able to determine that the weight ratio of oxygen to hydrogen is 8/1 in water, and because, in his formula for water, HO, there was one oxygen atom and one hydrogen atom, he assigned a relative weight of 8 units to the oxygen atom.

At the beginning of the nineteenth century, the French chemist Joseph Louis Gay-Lussac (1778–1850) was working on the reaction of hydrogen and oxygen in an effort to develop a method of determining the oxygen content of air. He mixed excess amounts of hydrogen with oxygen, igniting the mixture with a spark, and then measuring the change in volume (Garrett 1968, pp. 162–168). In this context, he performed experiments which showed how oxygen and hydrogen combined. Gay-Lussac noticed that two volumes of hydrogen combined with one volume of oxygen to form just two volumes of water vapour. In addition, he found that other gases also exhibited the same tendencies (Hurd and Kipling 1964, pp. 46–62; Mahan 1972; Partington 1960, pp. 180–215). Gay-Lussac explained this result as ‘when measured at constant temperature and pressure, the volumes of gases that are used or produced in a chemical reaction can be expressed in ratios of small whole

numbers' in 1808. Today, this statement is called Gay-Lussac's law of combining volumes (Gay-Lussac 1809).

Gay-Lussac claimed that the weights of equal volumes of gases in combination would be directly related to the combining weights and so to the atomic weights. He said that his results were consistent with both Proust's law of definite proportions and Dalton's law of multiple proportions about the chemical composition of molecules, and that his observations of combining gases in simple ratios by volume was empirical evidence for Dalton's idea of substances combining atom to atom. Also, he stated that his results were 'very favourable to Dalton's ingenious ideas' about the composition of molecules. However, Dalton did not agree with him. According to Dalton, there were problems in linking his ideas with Gay-Lussac's experiments. Dalton rejected the 'law of combining volumes' as an exact description of gas reactions (Garrett 1968, pp. 162–168). He said that Proust's law of definite proportions and his law of multiple proportions in chemical composition referred to combining weights and not to volumes. He saw that Gay-Lussac's observations implied that the space occupied by an atom was the same for all gases, and that the numbers of particles contained in equal volumes of gases were either equal or integral multiples of one another. However, at that time, Dalton was convinced that the number of particles in an equal volume of different gases could not be equal; he argued that the sizes of gas molecules were different enough to make it impossible to have equal numbers of molecules (Garrett 1968, pp. 162–168). Thus, the appearance of integer relationships, which seemed to Gay-Lussac to support the atomic theory, found the least favour with the father of atomic theory.

In 1811, Avogadro saw what Dalton and Gay-Lussac could not see. Dalton's atomic model and Gay-Lussac's observations on combining volumes would be mutually consistent if there were a simple relationship between atoms and volumes. Especially Dalton's atomic model would provide an excellent explanation of Gay-Lussac's observations if every one of Gay-Lussac's volumes contained the same number of Dalton's atoms.

Avogadro cleared up the difficulties for the application of the law of volumes. His starting point was the results of the earlier studies, and he interpreted these results on gases. In 1766, the British chemist Henry Cavendish (1731–1810) synthesized water from hydrogen and oxygen (Partington 1960, pp. 122–152). In 1800, two British chemists, William Nicholson (1753–1815) and Anthony Carlisle (1768–1840), found that two volumes of hydrogen for each volume of oxygen are produced by the electrolysis of water (Russell 2011). As mentioned above, in 1808, Gay-Lussac discovered that two volumes of hydrogen combine with one volume of oxygen to form water. Based upon the findings of these three scientists about water and the work of Gay-Lussac, Avogadro proposed his hypotheses.

Avogadro's principal contribution to chemistry was a paper in which he made two hypotheses. The first hypothesis was that equal volumes of all gases under the same conditions of temperature and pressure contain the same numbers of molecules, and the second was that some elementary gases were composed of two atoms (Garrett 1968, pp. 162–167). These two hypotheses today are known as Avogadro's Law.

Avogadro was able to combine Dalton's Law of Multiple Proportions related to the weights of atoms with Gay-Lussac's law of combining volumes. As a result of the first hypothesis, he predicted that gas densities are proportional to atomic weights. He applied his law to the determination of the relative masses of gas molecules (Avogadro 1811). In addition, starting from the idea that 'the relative number of molecules in a compound is given by the ratio of the volumes of the gases that form it,' which is a result of Gay-Lussac's law, Avogadro demonstrated that formulas could be predicted and molecular

weights could be computed in his theory. He deduced that the molecule of water contains half a molecule of oxygen and one molecule of hydrogen. He assigned a weight of 1 unit to the hydrogen atom, like Dalton, and calculated the relative atomic weight of oxygen as 16 mass units. However, the European scientific community, and especially Dalton, never accepted Avogadro's Law.

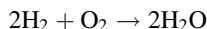
According to Dalton, since equal volumes of gases could not contain the same numbers of molecules, gas densities should not be directly proportional to atomic weights. Then, Avogadro's atomic weight scale could not be true. The most important issue was that Dalton could not see the difference between atoms and molecules, or perhaps did not want to understand it. If one volume of oxygen combined with two volumes of hydrogen to form two volumes of water, it looked to him as if each of the water molecules had half an oxygen atom. However, the divisible things in Avogadro's work were molecules, not atoms. Again, according to him, two atoms combined with each other due to electrical forces between them, and two atoms of the same element could not combine with each other because they would repel each other as all like charges do. Therefore, none of the elements could be polyatomic in nature and one molecule of water could not contain two hydrogen atoms (Garrett 1968, pp. 162–167).

Avogadro's hypotheses were overlooked for 50 years because the scientific community of the early nineteenth century did not know Avogadro well and could not appreciate his ideas. Thus, half a century of chaos existed in chemistry, due to the confusion about the distinction between atoms and molecules and the rejection of Avogadro's ideas.

In the first half of the nineteenth century, the problems of determining the atomic weight scale and molecular formulas became intensified. The permanent solution required only a slight extension of Avogadro's reasoning, and Cannizzaro provided what was needed (Cannizzaro 1858). In 1858, Cannizzaro came across the work of his countryman Avogadro. He saw how Avogadro's hypotheses could be used to distinguish between the atomic weight and the molecular weight of the important gaseous elements and how this distinction would serve to clarify the matter of atomic weights for the elements generally.

Based on Avogadro's second hypothesis and Gay-Lussac's law, Cannizzaro predicted the molecular formulas of hydrogen and oxygen gases as H_2 and O_2 , respectively. He assigned a weight of 1 unit to the hydrogen atom, like Dalton and Avogadro, and calculated the relative atom weight of oxygen as 16 mass units. Then, in the researcher's comment, in order to get the relationships among Dalton's atomic theory, Gay-Lussac's law of combining volumes and Avogadro's law, he wrote the following equations about the formation of water:

hydrogen + oxygen \rightarrow water



Dalton: $2N \times 2$ mass units hydrogen + $N \times 32$ mass units oxygen
 $\rightarrow 2N \times 18$ mass units water

Gay-Lussac: $2V$ volume units hydrogen gas + V volume units oxygen gas
 $\rightarrow 2V$ volume units water vapour

Avogadro: $2N$ molecules hydrogen + N molecules oxygen $\rightarrow 2N$ molecules water

Thus, Cannizzaro showed that these three scientists, in fact, also indicated the same things; he reconciled them to each other despite the fact that there were no!

But, how would these results about a subject which had been discussed for 50 years be explained to the European scientific community, and how would the community be persuaded? In 1860, the first International Congress of Chemistry in history was held in Karlsruhe, Germany, in order to clear up the confusion about atomic weights. One hundred and forty delegates, including Cannizzaro, attended the conference. At the Congress, he made a strong speech on the subject and then distributed copies of a pamphlet in which he explained his point of view about how Avogadro's hypotheses could resolve all these questions. Thus, Cannizzaro's efforts persuaded most of the other delegates and a consistent system of atomic weights was finally established. Cannizzaro's atomic weights became more and more usual and useful. A direct outcome was that the formal concept of the mole was founded on the firm ground of a stable system of atomic and molecular weights (Cerruti 1994; Partington 1960, pp. 240–272). With Avery's (2005, p. 186) expression, unfortunately, neither Dalton nor Gay-Lussac nor Avogadro saw the triumph of their theories and again, none of them heard a torrent of applause for their triumph at Karlsruhe since they were no longer living.

Avogadro, Loschmidt, Károly than and the number $6.022 \times 10^{23} \text{ mol}^{-1}$

Thus, Cannizzaro calculated the atomic weights of the known elements on the basis of the hydrogen atom. However, because individual atoms are far too small to weigh, the chemists' measuring unit needs to be very big. This unit is the mole. According to the research literature (e.g. Gorin 1994), the term 'mole' was introduced into chemistry by Ostwald with the sense of gram-molecular weight in 1900. Ostwald defined the mole as 'the amount of any gas that occupies a volume of 22,414 mL in normal conditions is called one mole.' In the following years, scientists needed to know the number of particles in this volume. Again, according to the research literature (e.g. Becker 2001; Giunta 2011b; Murrell 2001), Perrin calculated this number as $7.05 \times 10^{23} \text{ mol}^{-1}$ in 1908. Perrin named this number as 'Avogadro's number' due to following reason:

According to the first hypothesis of Avogadro's law, the equal volumes of different gases at the same temperature and pressure contained the same number of particles. If the equal volume at the same temperature and pressure of 0°C (273.15 K) and 1 atm (101.325 kPa) is 22.414 L (0.022414 m^3), it is the volume of one mole gas to be a result of Ostwald's definition of mole. Therefore, the number of particles in one mole gas is 7.05×10^{23} . Since this number was a result of Avogadro's law, it should be named as 'Avogadro's number.'

In fact, the number $6.022 \times 10^{23} \text{ mol}^{-1}$ was not discovered and calculated by Avogadro. He did not even imply such a number in his essay in 1811 (Avogadro 1811). However, since Perrin saw Avogadro to be an original creator of the idea of the number $6.022 \times 10^{23} \text{ mol}^{-1}$, he called this number 'Avogadro's number' to honour the man who never received recognition during his life for his substantial contributions to early chemistry 50 years after his death. Thus, Avogadro was acknowledged for the second time after the Congress of Karlsruhe.

It is clear that Avogadro made no quantitative estimate of the number $6.022 \times 10^{23} \text{ mol}^{-1}$. However, who calculated the number for the first time and when? According to Becker (2001), in 1865, Loschmidt for the first time estimated it as $72 \times 10^{23} \text{ mol}^{-1}$. Again, according to the same researcher, in 1908, Perrin obtained a more accurate value, $6.7 \times 10^{23} \text{ mol}^{-1}$, for Avogadro's number. The fact that Loschmidt was honoured as the first person who estimated this number was due to following reason.

In 1865, Loschmidt for the first time calculated the number of molecules in one cubic centimetre of a gaseous substance under ordinary temperature and pressure. In 1900, Ostwald defined the mole as 'the amount of any gas that occupies a volume of 22,414 mL under normal conditions' (Gorin 1994). In 1908, Perrin defined Avogadro's number as 'the number of molecules in one gram-molecular volume (namely 22,414 mL) of a gas under normal conditions' (Becker 2001). If the number which Loschmidt calculated is multiplied by 22,414 mL in Ostwald's and Perrin's definitions, the result is the number of molecules in the gram-molecular weight of any gas, and it is Loschmidt's number in the sense of the $6.022 \times 10^{23} \text{ mol}^{-1}$.

Alright! But, by whom and when was the number $22,414 \text{ mL mol}^{-1}$ determined for the first time? In the researcher's opinion, this knowledge is a keystone to be established for the first determination of the number $6.022 \times 10^{23} \text{ mol}^{-1}$.

Wisniak (2000) stated that Cannizzaro measured the value 22.4 L mol^{-1} . Also, in a document written by Gladney (1999), the following statement was given: the volume of a gas at normal conditions was predicted by Avogadro as 22.4 L per mole. These different expressions were even enough to show that neither Avogadro nor Cannizzaro calculated the number 22.4 L mol^{-1} . Above all, the concepts of the gram-molecular weight and gram-molecular volume and the number 22.4 L mol^{-1} were implied in neither Avogadro's nor Cannizzaro's articles since their focuses were not them. Their goals were to develop an accurate atomic weight scale and find a way to estimate the molecular formulas. Each was able to reach the goal without needing the concepts of gram-molecular weight and gram-molecular volume. Furthermore, at the time that the problems of determining atomic weight scale and molecular formulas were not yet solved, the determination of the gram-molecular volume or the molar volume should not be expected, since the concepts such as the mole, the gram-atomic weight, the gram-molecular weight, the molar volume and the gram-molecular volume are based on the atomic weight and the molecular formula. This case can be seen in Avogadro's (1811) and Cannizzaro's (1858) essays translated in the Alembic Club.

In 1865, Loschmidt first determined the number of molecules in a cubic centimetre of a gas under normal conditions to be 1.83×10^{18} (Hawthorne 1970). The concept of mole was first introduced into chemistry by Horstmann, as cited by Cerruti (1994) and Turco and Cerruti (2002) by the term 'gram-molecular weight' in 1881. Károly Than first determined the gram-molecular volume (molar volume) of gases at the temperature of 0°C and pressure of 1 atm as $22,330 \text{ cm}^3$ in 1889 (Than 1889). Accordingly, the first value of Avogadro's number which was calculated from these two numbers ($1.83 \times 10^{18} \text{ molecules/cm}^3$ and $22,330 \text{ cm}^3$) in the history of science was $4.09 \times 10^{22} \text{ molecules/gram-molecular weight}$. Although the number $4.09 \times 10^{22} \text{ molecules/gram-molecular weight}$ was a rough estimate compared to today's value of Avogadro's number, it represented the concept for the first time. The reason for this rough value was a large experimental deviation in Loschmidt's number.

In chemistry literature, it is sometimes expressed that the number $6.022 \times 10^{23} \text{ mol}^{-1}$ was estimated by Loschmidt for the first time (e.g. Becker 2001; Murrell 2001). Furthermore, in German literature, one can often find Avogadro's number referred to as Loschmidt's number per gram-molecule (Murrell 2001). However, Loschmidt calculated the number of molecules in one cubic centimetre of gaseous substance under ordinary conditions of temperature and pressure, not the number of molecules in 22,414 cubic centimetres. Here, one should understand the distinction between Loschmidt's number and Avogadro's number: Loschmidt's number is the number of molecules present in a cubic centimetre of a gas and Avogadro's number is the number of molecules present in 22,414

Fig. 1 Károly Than (1834–1908), Professor of Chemistry at the University of Budapest (1860–1908) (from László 2011)



cubic centimetres of a gas under normal conditions. This duality causes the confusion in chemistry. In addition, the fact that Than contributes to the first calculation of the number $6.022 \times 10^{23} \text{ mol}^{-1}$ is an evident case. In the researcher's opinion, this duality (also this wrong knowledge) in the naming of number $6.022 \times 10^{23} \text{ mol}^{-1}$ should be given an end and this number should be called only Avogadro's number. In addition, Than's contributions to its first calculation should be acknowledged.

Unfortunately, Than is not a well-known figure in the science world. Although the first definitions of the mole and the number $6.022 \times 10^{23} \text{ mol}^{-1}$ are based on the number 22,414 mL mol^{-1} , the question 'who is the first owner of this number' and an answer to it have been omitted for over a 120 years. Károly Than (Fig. 1) was the founder of modern Hungarian chemistry and he contributed to science and science education in Hungary as well as to chemistry in general with his views for more than half a century. For more detailed knowledge about Than, some related sources (e.g. Kauffman 1989; László 2011; Szabadváry 1966; Vámos 2006, 2007) should be read.

Conclusion

As expressed above, the foundation for the idea of the number $6.022 \times 10^{23} \text{ mol}^{-1}$ was constructed by Avogadro in 1811. Loschmidt and Than together made the first calculation of this number as 4.09×10^{22} molecules/gram-molecular weight in the years of 1865 and 1889. Horstmann, for the first time, introduced the concept of gram-molecular weight into chemistry in 1881. Ostwald used the word 'mole' instead of gram-molecular weight in 1900. Perrin calculated a more accurate value for the number of molecules in one mole and called this number Avogadro's number in 1908. Hence, Avogadro is the originator of the ideas of the mole and the number $6.022 \times 10^{23} \text{ mol}^{-1}$, and Loschmidt and Than are the scientists who first calculated Avogadro's number. Accordingly, the number 4.09×10^{22} molecules/gram-molecular weight, which is calculated by multiplying Loschmidt's 1.83×10^{18} molecules/ cm^3 by Than's 22,330 cm^3 /gram-molecular weight, today's accepted experimental value, $22.413\,996 \times 10^{-3} \text{ m}^3 \text{ mol}^{-1}$ (NIST 2011), should be

accepted to be the first value for Avogadro's number in science history. In addition, Than's contributions to its first calculation should be acknowledged.

The distinction between Loschmidt's number and Avogadro's number should well be understood: Loschmidt's number is the number of molecules present in a cubic centimetre of a gas and today's accepted experimental value is $2.686\ 7774 \times 10^{25}$ particles m^{-3} (NIST 2011), and Avogadro's number is the number of molecules present in 22,414 cubic centimetres of a gas under normal conditions, and today's accepted experimental value (the number of atoms of ^{12}C in 12 grams of ^{12}C) is $6.022\ 141\ 79 \times 10^{23}$ particles mol^{-1} (NIST 2011).

As an educational recommendation, Dalton's, Gay-Lussac's and Avogadro's stories should be told in science textbooks. Again, there should also be the short histories of the mole and the number $6.022 \times 10^{23} \text{ mol}^{-1}$ in science textbooks. The roles that particularly Cannizzaro, Loschmidt, Horstman, Ostwald and Perrin play in science are not completely known by the young generation, and also teachers, instructors, and lecturers, since their names do not usually appear in textbooks. Moreover, although Than is one of two scientists who calculated for the first time the number $6.022 \times 10^{23} \text{ mol}^{-1}$, he has hardly ever been recognized for about a 150 years. However, each of them (Dalton, Gay-Lussac, Avogadro, Cannizzaro, Loschmidt, Horstman, Than, Ostwald and Perrin) is a milestone in the science world. Therefore, their names should be in science textbooks in all level of education, which are sources which people can easily reach. In order to feel respect and admiration for these scientists, be aware of the difficulties which they experienced and better and more efficiently teach and learn science concepts, all these things should be done. According to the researcher's observations in classes, students like to study science history; they are affected by the life stories of scientists. Students who are bored by the traditional chemistry topics are all attention in the science history topics. The researcher always recognizes that his students' eyes moisten when he explains that Avogadro died without learning the fact that his law had been acknowledged. That is, if the science history and/or philosophy knowledge are given, they are ready to take. In addition, the researchers (Adúriz-Bravo 2004; Erduran et al. 2007; Freire and Tenório 2001; Giunta 2001; Harrison 2002; Lin et al. 2002; Matthews 1989, 1992, 1994, 2007; Quílez 2004; Sarikaya 2007; Seroglou and Koumaras 2001; Watson 2007) have always indicated the important of using the history, nature and philosophy of science to better teach science, and emphasised that the science curriculum should include the history, nature and philosophy of science.

The essence of the present study is the emphasises that Horstmann first introduced the mole concept into science/chemistry and the first value of Avogadro's number in the history of science was 4.09×10^{22} molecules/gram-molecular weight and Loschmidt and Than together first calculated this number, and especially the fact that the value 4.09×10^{22} molecules/gram-molecular weight was the first value of Avogadro's number in science/chemistry history and Károly Than's role to calculate this number are first emphasised in the present study.

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