Anal Bioanal Chem
DOI 10.1007/s00216-011-4908-4

FEATURE ARTICLE

Origin and early history of *Die Methode des Eichzusatzes* or *The Method of Standard Addition* with primary emphasis on its origin, early design, dissemination, and usage of terms

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The Method of Standard Additions or The Standard Addition Method, often referred to by its acronym as just SAM, is a proverbial workhorse in both inorganic and organic quantitative analytical chemistry and in related disciplines such as geochemistry. Its advantage in mitigating the effects of matrix interferences compared with the calibration curve approach is well known and is one of its major benefits. It is presented in virtually all standard textbooks on analytical chemistry in varying degrees of complexity. Yet the story of how it originated, and by whom, is not well known. It is generally believed that it originated in the early 1950s, introduced by spectroscopists. We have determined that the priority of its use and discovery apparently belongs exclusively to Hans Hohn (1906–1978), a mining chemist, dating from the exposition of the method in his 1937 book on polarography, *Chemische Analysen mit dem Polarographen*. How the method became established in other disciplines quite different from polarography is not completely clear. It is likely that it was *rediscovered* independently in at least two separate disciplines, X-ray fluorescence (XRF) and atomic spectroscopy, almost simultaneously in 1953, 16 years after Hohn's original description.

Dedicated to Hans Felix Ferdinand Julius Richard Hohn, the originator of The Method of Standard Addition

Electronic supplementary material The online version of this article (doi:10.1007/s00216-011-4908-4) contains supplementary material, which is available to authorized users.

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Die Methode des Eichzusatzes or literally "the method of calibration addition" was first used and described by Hans Hohn [1] in 1937 in his book *Chemische Analysen mit dem Polarographen* (see supplementary material), a practical book with 28 experiments followed by nine detailed "examples of polarographic analyses". Polarography had been invented by Jaroslav Heyrovský in 1922 [2] and he and colleague Masuzo Shikata described the first automatically recording instrument in 1925 [3]. Heyrovský received the 1959 Nobel Prize in chemistry "…for his discovery and development of the polarographic methods of analysis" [4] which was the first Nobel for a chemical method of analysis. Polarography was a very popular and much used technique well into the 1960s until the advent of modern spectroscopy. The literal English translation of ***Eichzusatzes*** into ***calibration addition*** comes from joining the verb, *eichen*, and the noun, *Zusatz*, which mean *to calibrate* and *addition*, respectively (***Eichzusatz*** – singular; ***Eichzusätze*** – plural; ***Eichzusatzes*** – genitive of the singular). Hohn's new book on polarography was very favorably reviewed by Samuel Glasstone in 1938 [5] who noted that Hohn was "…associated with a famous smelting works in Duisburg…." Glasstone, showing a scientist's unique sense of humor, stated in closing, "…the polarograph sometimes behaves in a manner unaccountable to the beginner" and therefore having this book "…will save him

Published online: 21 April 2011

both time and anxiety." The title page Hohn's [1] book is reproduced in Fig. 1.

Later in the German literature the common description of the method became ***Die Methode der Standardzugabe*** which literally means the Method of Standard Addition (*Zugabe* = addition) or ***Standardzugabemethode***. For example, Schejtanow [6], in Bulgaria titled his 1957 paper *Über die Genauigkeit der polarographischen Methode der Standardzugabe*. See also the paper by Pliška [7] from 1962 on uncertainty analysis for calibration, internal standards, and *Standardzugabemethode*. Another variation was ***Eichzusatzmethode*** [8, 9]. It is important to note that the word *addition* is always singular in the polarographic literature. There are more than 50 references to Hohn's book in the research literature and numerous references in the early polarographic and electroanlytical textbooks. All the evidence that we have examined suggests that the priority for the use and description of the *Method of Standard Addition(s)* belongs exclusively to Hans Hohn as subsequent discussion and references will demonstrate. It was used in polarography for almost 20 years before it was used in X-ray fluorescence (XRF) and atomic spectroscopy.

Perhaps the earliest reference book outside the field of polarography to advocate the Method of Standard Additions is *Spectrochemical Procedures* by Harvey [10], an obscure book published in 1950 by his employer the Applied Research Laboratory (ARL). Harvey devotes essentially the whole of Chapter IX to the "*Addition Method of Analysis*" without attribution. In his presentation the axes are reversed, the addition axis is the ordinate and the response axis is the abscissa. The often stated idea for the last 50 years that SAM is an extrapolation method was not a legacy of the polarographers, but rather later spectroscopists. Harvey may have been the first to refer to the addition method as an extrapolation method, an idea that gained universal acceptance and is still prevalent today, but was recently challenged by Kelly and Guthrie [11]. Harvey's graphical presentation and description as "*Addition Method of Analysis*" did not gain acceptance. The method is now most commonly referred to as the *Standard Addition Method*, the Americanized noun-adjective form, and frequently by the acronym ***SAM***.

Figure 2 is an illustration of a common design of SAM experiments. In this design successive additions were made to five samples (X_1 to X_5) with no addition to sample X_0. The estimated straight line fit to the data was determined by weighted least-squares regression. The concentration may be read from the geometrical extrapolation to the *X*-axis at *Y*=0 which has led to the common belief that SAM is extrapolation.

Surprisingly, this well known design bears no resemblance to the original ***Eichzusatz*** design of Hohn and subsequent polarographers who developed it. Hohn's contrived example of the concept of standard addition is reproduced in Fig. 3. The figure shows the polarogram of the original solution, labeled a, containing known amounts of Cu, Tl, Zn, and Mn. After addition of a known amount of Zn to the original solution a second polarogram, labeled b, is obtained. The Zn response has increased from

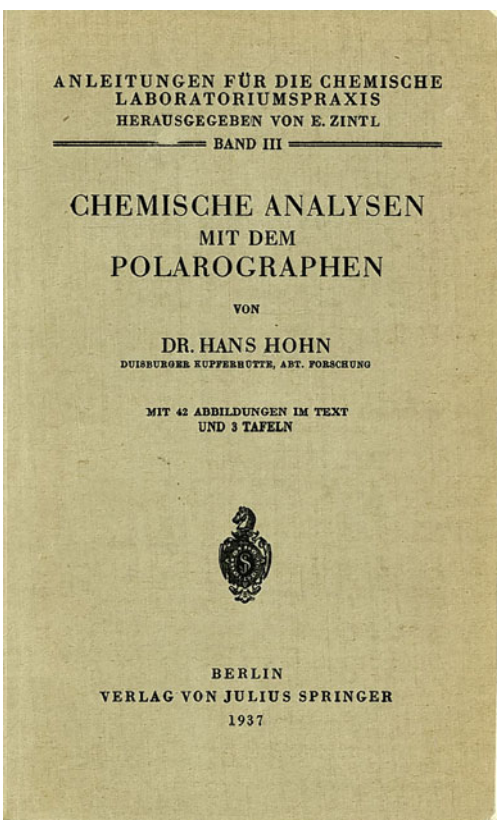
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Fig. 1 Cover of Hohn's book on polarography containing the first description of The Method of Standard Addition

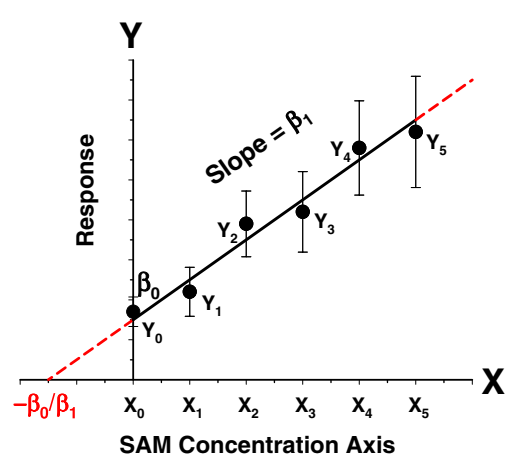

Fig. 2 Traditional SAM plot with five additions of increasing amounts (X_1 to X_5) and one sample without addition at the origin of the abscissa (X_0, Y_0). The increasing length of the uncertainty bars of the Y_i responses with the X_i values indicates heteroscedastic data

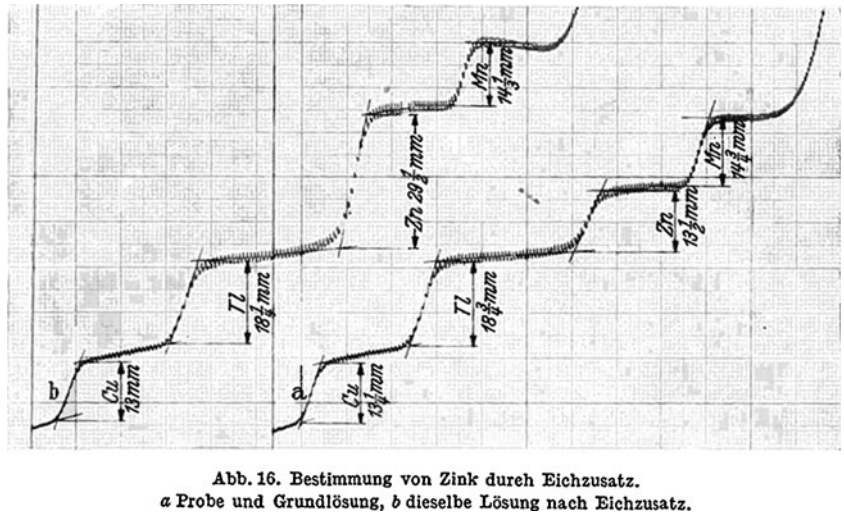

Fig. 3 Polarogram of Zn standard addition experiment from Hohn (Fig. 16). The caption reads, "Figure 16. Determination of Zn by calibration (standard) addition. a Unknown and supporting electrolyte, b same solution after calibration addition"

13.5 mm to 29.5 mm whereas the response of the other cations is unchanged. The amount of Zn in the original solution may be calculated using Hohn's exact formulation:

$$x = \frac{A \cdot m \cdot ccm_{+} \cdot ccm_1 \cdot ccm_2 \cdot W_1}{1000 \cdot ccm_2 \cdot (W_2 \cdot ccm_2 - W_1 \cdot ccm_1)} \quad (1)$$

A	is the atomic or molecular weight of the sought substance;
m	is the molarity of the added solution
ccm_1	is the volume of the original solution
ccm_+	is the added volume of calibrating solution
ccm_2	is the final total volume
W_1	is the original wave height
W_2	is the final wave height; and
x	is the searched-for amount in the sample, in grams.

The appearance of the term ccm_2 in both the numerator and denominator is Hohn's and is mathematically correct. Hohn's equation yields the absolute amount of the unknown substance in grams, but can be easily rearranged to yield concentration by dividing both sides by ccm_1 (volume 1 in cubic centimeters). The latter form is the formulation used in most subsequent publications. Figure 3 may well be the first published example of *Die Methode des Eichzusatzes*. On page 84 Hohn gives a recipe for determination of the contaminants Cu, Pb, Zn, Fe, and In in pure aluminium sheet. The sample was dissolved in hydrochloric acid, taken to near dryness and brought to a known volume by addition of water. In this case the supporting electrolyte was the aluminium chloride solution itself. Hohn then says that it is best to use an *Eichzusatz* for these determinations.

In the first complete review article on polarography published in 1939 (manuscript submitted December 12, 1938), Kolthoff and Lingane [12, 13] reference Hohn's [1] book 13 times, and give a very clear description of SAM:

> "The polarogram of the 'unknown' solution is first obtained in the usual way. A known amount of the substance in question is then added to the solution, and a second polarogram is obtained. From the increase (corrected for the residual current) of the diffusion current caused by the known addition, the original concentration of the substance can be easily be computed by simple proportion. The particular advantage of this method is that the 'calibration' is obtained under identically the same conditions with which the original polarogram was obtained…"

This statement completely captures the essence of SAM:

- at least two measurements are required, the first without and the second with addition of the element of interest; and
- the sample itself is used to construct the calibration curve and thus the measurement conditions are the same except for the perturbation of the analyte concentration by the addition of the analyte of interest.

The two-level design described by Kolthoff and Lingane was the design originated by Hohn and subsequently used by all early polarographers. This is the reason that polarographers always refer to the method in the singular, standard addition or *Eichzusatz*, whereas the spectroscopists, who use successive dilutions (Fig. 2), always use the plural, standard additions.

The first use of the term "standard addition" in a research paper in English is by Lingane and Kerlinger [14] in 1941 in *Analytical Chemistry* on the determination of Ni and Co by polarography where they define Δi as equal to the "increase in diffusion current resulting from standard addition". In the same year three books were published on polarography and all mention The Method of Standard Addition. The first, *Polarography*, was the first in English written by Kolthoff and Lingane [15] (Forward

dated March 15, 1941) "…a book so well written that it has been called the 'polarographic bible' " [16]. It has a section, "Method of Standard Addition," but it is not listed in their subject index, not an infrequent occurrence in early textbooks on the subject. They state that "This method… appears to have been originated by Hohn" and they reference Hohn's book [1] and the earlier paper of Lingane and Kerlinger [14]. In total there are nine references to Hohn's book. An entry for "standard addition calibration technique" was added to the subject index of the 2nd addition [17] of 1952, but the discussion of SAM is essentially the same as that in the 1st edition. This was followed by Heyrovský's monumental work [18] *Polarographie* (Forward dated October 1941), a comprehensive 514-page work in German based on, but not directly translated from, his earlier book [19] in Czech. Heyrovský presents the mathematical formulation on pages 265–267. This was followed a few months later by a small book by Otto Müller [20] based on a series of five articles he had written for the *Journal of Chemical Education*, also in 1941. Müller had been a visiting researcher in Heyrovsky's laboratory between 1933 and 1935.

The last two books do not mention standard additions specifically, but both contained the mathematical formulation for the two-level standard additions procedure without attribution. Both books list Hohn's book [1] in their references, but not in their mathematical formulations. Is it possible that Heyrovský may have formulated the concept before Hohn? The only evidence for this is found in his 1941 book which could have been based on earlier work published in the Czech language. There is some compelling circumstantial evidence to the contrary. Heyrovský published two exhaustive review articles in German in 1936 [21] and 1939 [22]. There is no mention of standard additions in either. In the first, as expected, there is no mention of Hohn's 1937 book. In the second he cites both Hohn's 1935 [23] short article and Hohn's book [1] in the literature section under the listing, "review papers" (*Zusammenfassende Arbeiten*). In 1947, Taylor [24] had a section entitled "Methods of Standard Addition" in a paper on *Examination of Absolute and Comparative Methods of Polarographic Analysis*. His citations for the method are Lingane and Kerlinger [14] and a 1946 paper by Cranston and Thompson [25]. This latter reference is a paper on the determination of trace Cu in milk, and the authors used the terms "additive standard technique" and "Method of Standard Addition" and give their reason. They note that "…the amount of copper recovered was erratic. This condition was corrected by using an internal standard or ***'standard addition' technique as described by Hohn*** [5]" (emphasis added), a reference to Hohn's book [1]. Of note is Cranston's address, Laboratory of Polarographic Analysis, Chicago, an indication of the important status of the method in 1946.

In the 2nd edition of Müller's [2] book on polarography in 1951 there is a small section entitled "Method of Standard Additions" on page 150 without references in which he points out that this method "…is especially advantageous for isolated determinations". However, in the general reference section after three references to Jaroslav Heyrovský, his fourth reference is to Hohn [1]. The formulation presented is identical to the two-level presentation of Hohn [1]. The diffusion current from the sample with no addition (I_1) is measured and then the addition of n grams of C is added to m grams of the unknown and the second diffusion current (I_2) is measured and the unknown concentration, X, is determined from eq.(4) from the combination of eqs.(2) and(3) by division.

$$I_1 = kX \tag{2}$$

$$I_2 = \frac{k[mX + nC]}{m + n} \tag{3}$$

$$X = C\frac{nI_1}{I_2(m+n) - mI_1} \tag{4}$$

Equation (4) is identical to Hohn's original formulation if it were written in terms of concentration.

In 1955 Louis Meites [26] published a book on *Polarographic Techniques,* and stated in the preface (p. vii), "…as late as 1937 there were fewer than a dozen polarographs known to be in use in the entire United States." On page 178 he discusses SAM in Section 3 without attribution. He notes (p. 179), "Standard Addition Methods suffer from the practical disadvantage of considerably increasing the time required to complete the analysis. On the other hand, no other method is capable of greater accuracy under any conditions…" The next year Meites stated in a letter to Analytical Chemistry [27], entitled " '*Standard Addition' Method of Polarographic Analysis,*"

> "One of the most valuable techniques of quantitative polarographic analysis is the Standard Addition Method ***devised by Hohn*** [1] and discussed by Kolthoff and Lingane [2], Taylor [4], and Meites [3]" (emphasis added, reference numbers are those of Meites and correspond to Refs. [15], [24], and [26] herein).

Finally, one last attribution of SAM to Hohn is given by Schejtanow [6] in his 1957 paper on the accuracy of polarography by The Method of Standard Addition. Schejtanow credits Hohn for the *Methode der Standardzugabe* in the opening line.

In summary, many attributions of SAM are given to Hohn's description in his 1937 book [1], and there are no references or attributions to anyone else. Hohn apparently

published only one paper prior to his book—a single column paper in 1935 [23] with the same title as his forthcoming book. There is no mention of SAM in this paper and the date in the forward of his book is given as January 1937, suggesting that he may have come to the idea of standard addition between 1935 and late 1936.

Polarographers used a two-tier design in all their early papers and determined the unknown value by simple proportion. Therefore, they did not need or use any type of regression analysis and never mentioned it. Meites [27] presented the first uncertainty analysis for the two-level SAM approach. Again Lingane [28] in the 2nd edition of his landmark textbook in 1958, *Electroanalytical Chemistry,* presents eq. (4), discusses the method without naming it, and omits it in the subject index. It is likely that this section would have appeared in his first edition [29] in 1953, but this edition did not contain a section on polarography, as he explained in the preface. In 1957 Porter [30] stated that "…the most accurate results in polarographic analysis were obtained by careful Standard Addition Methods". It would appear that the two-level SAM design was well entrenched in polarography well into the late 1950s. It was only later in 1966 in the work of Kemula [31] and Kemula and Sacha [32] that successive dilutions were used in polarography. Kemula [31, 33] described a novel "sluice" system for injection of the standard.

In 1968 Heyrovský and Zuman [34] published a practical book on polarography intended for students. This book is based on earlier works and is basically an integration of all work on polarography before 1968. The subject index contains six references to the "Standard-Addition Method" but there is no reference to Hohn [1]. This is a surprising omission considering the encyclopedic knowledge of the authors and the extensive and comprehensive references to the polarographic literature. The polarographic literature that we have examined attributed SAM exclusively to Hohn through 1959 [35].

With the exception of the book by Harvey [10] mentioned earlier, the first description and use of SAM beyond the field of polarography was by Campbell and Carl [36, 37] in 1954 (Table 1). They used SAM in the determination of niobium and tantalum in ores by "fluorescent X-ray analysis," a newly-developed technique, subsequently referred to as X-ray fluorescence spectrometry (XRF). Indeed, the authors state, "The first commercially available model of a fluorescent X-ray spectrograph, manufactured by North American Phillips Co. has been used in these studies." They present three different techniques for these determinations, the third one, their Method C, being "…an additive technique". They point out, "If too little oxide is added, experimental errors may be large. If the amount added is too large, the approximation of a linear relationship is poor". They suggest that $I_x / I_{x'+y}$ should be between 0.7 and 0.8. They then make two observations that became intrinsically associated with SAM,

> "If high accuracy is required, the sample may be analyzed by a series of ***successive dilutions***. The results are then ***extrapolated*** back to zero addition to obtain the best answer" (emphasis added).

This paper is noteworthy because it is the first description of the idea of ***successive dilutions*** and the first use of the word ***extrapolation***. Spectroscopists, who applied SAM in their designs starting in the mid-1950s used equally spaced points, typically 5 or 6, as shown in Fig. 2. The final mathematical relationship given by Campbell and Carl [37] is a bit obtuse because they are adding a pure solid compound to the unknown sample and not a dilute solution of the analyte. Their equation can be shown to be identical to Hohn's [1].

Now comes a bit of a controversy with regard to priority for SAM. It is commonly believed that the 1955 paper by chemical oceanographers Chow and Thompson [38] is the first description of SAM in the research literature and is frequently cited as such. Indeed, two of us recently made this mistake [39, 40] relying on "common knowledge" of our colleagues. The papers of Menis et al. [41] of the National Bureau of Standards (NBS, now NIST) from 1958 and Carr [42] from 1970 also attribute priority to Chow and Thompson [38]. This is probably because Chow and Thompson were the first to use SAM to determine an analyte in a complex natural

Table 1 Chronology and dates of two early publications of SAM in spectroscopy

Chronology of publication	Dates and citations	
	Campbell and Carl [37]	Chow and Thompson [38]
Table V. Oldest ocean seawater sample	–	7 July 1952
Presented at Pittsburgh Conference [38]	2–6 March 1953	–
Received for review	24 September 1953	14 June 1954
Accepted for publication	22 January 1954	5 October 1954
Published	May 1954	January 1955
Analytical Chemistry citation	1954 Volume 26 No. 5	1955 Volume 27 No. 1
Number of citations (September 2010)	33	73

aqueous medium, Sr in seawater, and SAM is most often used with aqueous solutions.

The Campbell and Carl [37] and the Chow and Thompson [38] papers were published eight months apart in *Analytical Chemistry* in May 1954 and January 1955, respectively. Table 1, gives the chronology of events. It is likely that both groups were operating independently of each other because of the closeness in submission dates and the fact they were in very different disciplines. Not surprisingly, neither references the other. Chow and Thompson [38] refer to the method as an *"internal standard"* method, and they used the idea of proportions to obtain their answer. Specifically, they state, "Since the graph…is a straight line, $Y = a + bx$, the unknown concentration, x, is given by the ratio of Y to (sic) slope of the line, when $a = 0$." They give the unknown concentration as equal to the intercept divided by the slope in their Fig. 2.

It seems from Table 1 that Campbell and Carl [37] were ahead of Chow and Thompson [38] in presentation and publication by almost two years (March 1953–January 1955). It is possible that Chow and Thompson could have started using SAM as early as the late summer of 1952, but not sooner, on the basis of the date of their oldest seawater sample. They state however, "Considerable time elapsed before the analyses of strontium were made" (p. 20 of Ref. [38]).

We made a completely unexpected and surprising discovery regarding the paper by Chow and Thompson [38]. It was stated above that they and Campbell and Carl [37] may have ***rediscovered*** SAM independently and almost contemporaneously in 1953–1954 as documented in Table 1. Because all the evidence points to Hohn and his 1937 book as being the first use and description of SAM, we wanted to examine Hohn's book in detail. Through inter-library loan, we received a copy of the book unexpectedly from the University of Washington in Seattle. The Chow and Thompson paper was submitted from the Department of Oceanography, University of Washington. Thomas G. Thompson (1888–1961) founded the oceanography department at Washington. Tsaihwa J. Chow (1924–2006) was one of his graduate students earning his Ph.D. there in 1960. Chow then spent his entire career at Scripps Institution of Oceanography. The copy of Hohn's book that we obtained still retained the circulation card inside the back cover, and the date-due slip. These items are reproduced in Fig. 4. Professor Thompson checked out this book on June 14, 1939, and was subsequently sent a notice three months later on 15 September 1939. Much later T. J. Chow checked out this book on November 26, 1951. Thus both Thompson and Chow had in their possession in 1939 and 1951, respectively, this very copy of Hohn's 1937 book describing standard addition, in each case before their 1955 paper in *Analytical Chemistry*. The omission of a reference to Hohn by Chow and Thompson may have been a result of their belief that they had extended the method of Hohn to such a degree that attribution was unnecessary. Hohn's method was a two-level design that could be solved by simple proportion. Chow and Thompson used multiple additions; their Fig. 2 shows a straight line defined by five data points essentially identical to our Fig. 2. They also presented extensive data on cation interferences in the determination of Sr. In a subsequent paper on the "Determination of Rb in Seawater" [43], of which Chow was a co-author, the paper of Chow and Thompson [40] is cited in connection with the "standard additions technique".

It is now re-established that Hans Hohn was the first to devise ***Die Methode des Eichzusatzes*** as a practical method to determine metals in complex solutions using the polarograph. To Hans Hohn alone goes the credit for the initial development and the algebraic formulation to determine the unknown concentration that remained essentially unchanged for almost 20 years. SAM remained exclusively within the polarography community until about 1950. The 1950 book by Harvey [10] is the earliest description by a spectroscopist. Thereafter it became a method of choice for spectroscopists who used multipoint linear least squares regression to determine the unknown concentration from the ratio of the regression parameter estimates. This practice continues today, essentially unchanged. One of the frequent advantages mentioned of SAM is that if one needs only to perform a single, or occasional, analysis, then SAM may be more time-efficient because one need not perform the necessary laboratory work to construct a multipoint calibration curve.

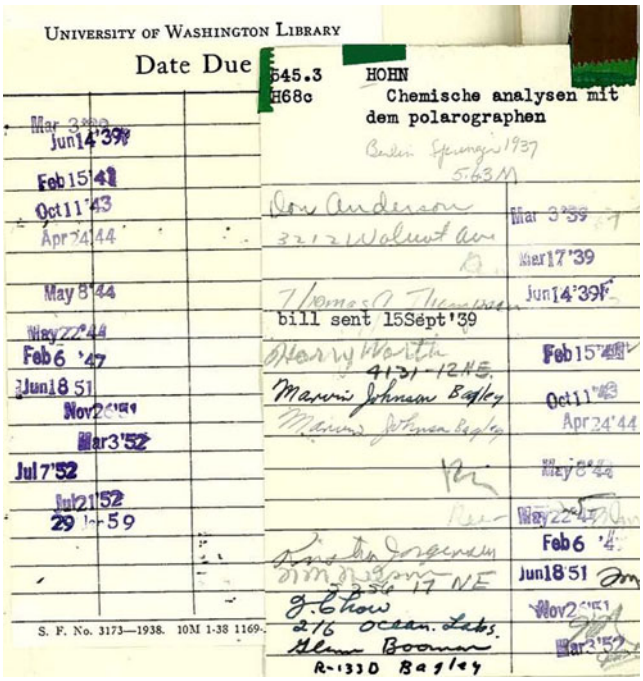

Fig. 4 Circulation card and date-due slip from the copy of Hohn's book in the library of the University of Washington

A more significant advantage is that the analyst need not know the matrix composition of the unknown sample.

It is of interest that polarographers used SAM extensively, but always using a two-level design, until the work of Kemula [31–33], and therefore never needed to resort to regression analysis. It is not clear how knowledge of The Standard Addition Method jumped from the community of polarographers into the disciplines of XRF and atomic spectroscopy. Harvey [10], Campbell and Carl [37], and Chow and Thompson [38] do not cite any other work in their discussions of SAM.

The tone of the descriptions of SAM in each of these references suggests that each group believed their formulation of the problem was original with them. It was true then and is still true now that the disciplines of polarography, XRF, and flame and plasma spectroscopy are completely independent, although researchers in these fields do publish in some of the same journals. The lesson for all scientists is that one should at least glance through or scan papers in completely different fields: one may find a useful idea.

Our conclusion is that all the evidence points to Hohn alone as the first to use and the first to describe the SAM, in 1937. Hohn's work was referenced by several subsequent workers in polarography. An obscure 1950 work by Harvey [10] was the first mention of the use of SAM in spectroscopy. In 1954, Campbell and Carl [37] seem to have discovered SAM independently. The impact of the Campbell and Carl paper is difficult to judge. Based on the citations in Table 1, it would seem that the subsequent Chow and Thompson paper had a greater impact. At the least, the Chow and Thompson work probably served to jump-start the use of SAM in the spectroscopic community. It would also seem that none of these groups realized the full impact of their work. None of them wrote a paper on SAM itself, but rather only used it. All three provided a theoretical basis for the method. Hohn [1] neither presented the method as new or novel, nor did he claim priority as he could have.

Acknowledgement The authors thank our NIST colleagues John R. Sieber for bringing Ref. [39] to our attention, Robert D. Vocke for suggesting editorial changes and helping with embedding the three figures from Hohn's book, and David L. Duewer for critical comments.

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40. The opening statement was, "The frequently used standard addition method (SAM) was first described for flame photometric analysis of Sr in seawater by Chow and Thompson more than 50 years ago." As stated, this assertion may be technically correct, but we do not wish to excuse our misattribution. We also inadvertently gave the wrong date of their paper as 1951 instead of 1955. We welcome this opportunity to correct the record.

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