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Paul Hagenmuller's contribution to solid state chemistry: A scientometric analysis

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ABSTRACT

Paul Hagenmuller (1921–2017) is an important figure of French solid-state chemistry, who enjoyed scientific and institutional recognition. He published 796 papers and has been cited more than 16,000 times. This paper explores Hagenmuller's work using scientometric analysis to reveal the impact of his work, his main research topics and his collaborations. Although Hagenmuller was a recognized scientist, a subset of his work, now highly cited, attracted little attention at the time of publication. To understand this phenomenon, we detect and study papers with delayed recognition, also called 'Sleeping Beauties' (SBs). In scientometrics, SBs are publications that go unnoticed, or 'sleep' for a long time before suddenly attracting a lot of attention in terms of citations. We identify 7 SBs published between 1965 and 1985, and awakened between 1993 and 2010. The first SB reports the discovery of the clathrate structure of silicon. The second reports the isolation of four new phases with the formula Na_xCoO_2 (x < =1). The five other SBs investigate the electrochemical intercalation and deintercalation of sodium, and the structure and properties of layered oxides. Through interviews with his coworkers, we attempt to identify the reasons for the delayed recognition and the context of the renewed interest in those papers.

1. Introduction

1.1. Paul Hagenmuller

Paul Hagenmuller (1921–2017) was a well-known and influential figure in the French solid-state community. He contributed heavily to the flourishing of a national academic community of solid-state chemists in post-war France, as well as the establishment of solid-state chemistry as a sub-field of materials research, with its own laboratory practices and its own objects of study [1]. Hagenmuller enjoyed scientific and institutional recognition in his country as well as internationally for his contributions to solid-state chemistry. For instance, between 1970 and 1975 he became a member of three German Academies of Science (Gottingen, Leopoldina, East Germany) as well as the Soviet Science Academy [2]. In addition to his achievements as a researcher, Hagenmuller trained many students who later became well known chemistry researchers.

Hagenmuller started a PhD thesis in 1945. His PhD supervisor was André Chretien, a professor at the Laboratory of Mineral Chemistry at the Sorbonne University in Paris. Meanwhile, he worked as a research assistant at the CNRS from 1945 to 1948, then as an assistant at the faculty of science in Paris from 1949 to 1950. After completing his PhD in 1950, he was appointed as a research fellow at the CNRS until 1953. He then spent two years in Vietnam as an associate professor in chemistry [2]. In 1956, Hagenmuller returned to France, and became an associate professor of mineral chemistry at the faculty of science in Rennes. This new academic position gave him scientific autonomy, which enabled him to recruit and train numerous PhD students, to increase his research efforts, and to gain a good reputation nation-wide. In 1961, the dynamism of this young professor led the University of Bordeaux to recruit him as well as most of his PhD students [3]. In 1964, Hagenmuller organized an international conference in Bordeaux on oxide crystals that is seen by some as the founding event of French solid-state chemistry [1]. In 1974, his laboratory – of which he was the director until 1986 – became “the Laboratory of Solid State Chemistry” thus marking the institutionalization of solid-state chemistry in France. The same laboratory became the *Institut de la matière condensée de Bordeaux* in 1995.

Throughout his career, Hagenmuller developed an approach to solid-state chemistry research that consisted of working at the intersection between chemistry and physics by studying the correlations between composition, structure, and properties of materials with the goal of creating new ones.

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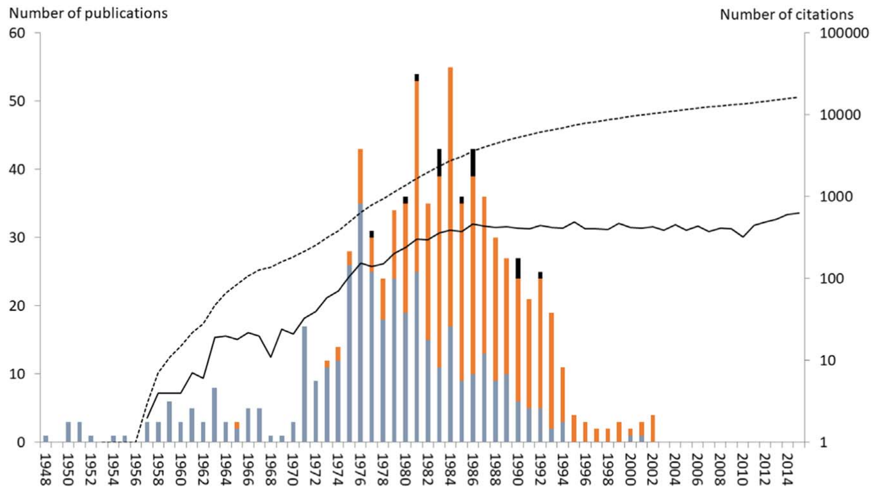

Fig. 1. Hagenmuller publications and citations. *Stacked columns,* number of Hagenmuller publications by year: *Blue*, publications in French, *Orange*, publications in English, *Black*, other languages; *black line,* citation distribution for all of Hagenmuller's publications; *dashed line,* cumulative citations for all of Hagenmuller's publications by year (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
Top 10 Journal distribution of Paul Hagenmuller's scientific production.

Source Title	Number of publications
Materials Research Bulletin	172
Journal of Solid State Chemistry	110
Comptes Rendus de l'Académie des Sciences	90
Solid State Ionics	40
Revue de Chimie Minérale	39
Solid State Communications	38
Journal of Physics and Chemistry of Solids	29
Zaac Journal of Inorganic and General Chemistry	26
Journal of the Less-Common Metals	23
European Journal of Inorganic and Solid State Chemistry	17

1.2. Scientometrics

Scientometric analysis is a powerful tool for studying scientific production quantitatively. As one of the most active scientific disciplines today, chemistry has been the subject of several scientometric investigations. Scientometric analysis was used to investigate specific chemistry topics [4], or to study chemistry-related issues at different levels: country [5], journal [6], or individual [7].

Scientometric analysis offers a panel of tools for studying the properties of a body of publications. Co-author analysis is useful for studying collaboration [8], and co-word analysis is used to explore the content of publications and identify research topics as well as their relationships [9]. Moreover, bibliographic coupling can be used to identify research fronts [10]. In addition, studying the citation patterns of a set of papers (e.g. highly cited papers) can indicate the durability of specific topics (i.e. the aging of scientific production) [11,12]. In particular, it is possible to characterize and study delayed recognition by identifying sleeping beauties (SBs) [13–15]. In scientometrics, a SB is a paper that does not achieve recognition in terms of citations until a few years after its original publication (it 'sleeps') [16].

Identifying and studying SBs can be of huge interest in order to understand the evolution and dynamics of scientific production. For instance, Ke et al. [17] found that "premature" topics can fail to attract community attention even when they are introduced by scientists who are highly recognized in their field, and that SBs tend to belong to clusters of publications that introduce such premature topics. In addition, Van Raan [18] found that a large proportion of SBs are application-oriented and can thus be seen as sleeping innovations.

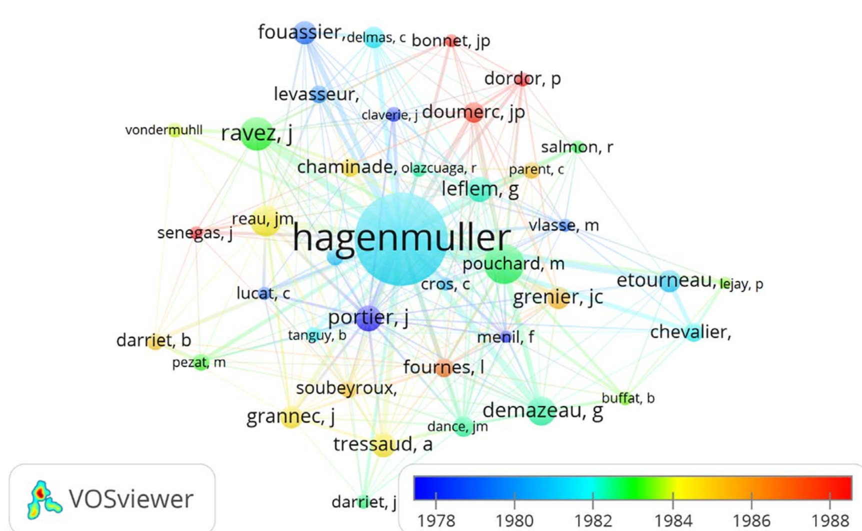

Fig. 2. Hagenmuller coauthors network through time (minimum 15 coauthored papers).

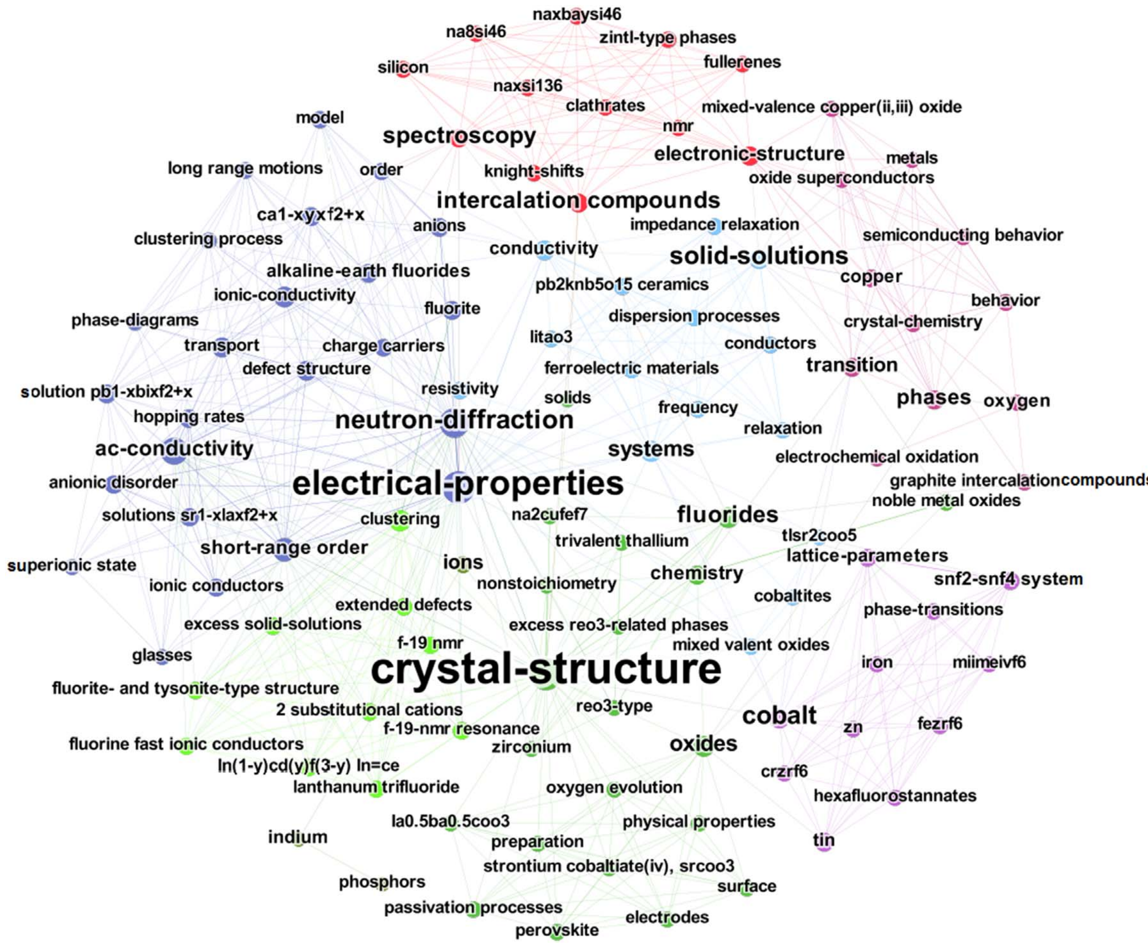

Fig. 3. Publication co-word network.

Table 2
Hagenmuller's main topics of research.

Cluster	Number of Publications	Main topics of research in the cluster	Main co-authors in the cluster
1	85	● stabilization of unusual, high oxidation states of transition metals under high pressure conditions ● Perovskite-type materials	Demazeau G; Grenier JC; Pouchard M
2	78	● Ferromagnetism in fluorides ● Magnetic properties of delafossite-type compounds	Dance JM; Grannec J; Tressaud A
3	67	● Ferroelectric and ferroelastic behavior in organic oxyfluorides	Chaminade JP; Pouchard M; Ravez J
4	65	● Fluorite-type solid solutions	Lucat C; Portier J; Reau JM;
5	61	● Nasicon materials ● Luminescence of rare-earth ions	Fouassier C; Leflem G; Parent C
6	57	● Crystal structure, magnetic and electric properties of various vanadium compounds	Casalot A; Demazeau G; Pouchard M
7	55	● Structure and properties of rare-earth borides	Chevalier B; Etourneau J; Lejay P
8	42	● Lithium-ion conductivity ● Structural classification and properties of the layered oxides ● Isolation and study of Na_xCoO_2 (x < =1) bronzes ● Intercalation chemistry	Braconnier JJ; Delmas C; Fouassier C
9	38	● Compounds with delafossite-type structure ● Photoelectrolysis of water	Dordor P; Doumerec JP; Pouchard M
10	36	● Ferroelectric materials	Bonnet JP; Ravez J; Vondermuhll R
11	35	● Ionic conductivity in glasses	Cros C; Reau, JM; Levasseur A
12	24	● Clathrate-type compounds ● Graphite intercalation compounds ● Mossbauer resonance investigations on the SnF_2-SnF_4 system, the SrF_2-SnF_2 system, the oxyfluoride Sn_2OF_2, etc.	Fournes L; Grannec J; Tressaud A
13	22	● The hydriding and dehydriding reactions of magnesium	Darriet B; Pezat M; Song MY
14	14	● Sillenite-type phases	Devalette M; Graciet M; Mazeau C
15	12	● Preparation of oxides with high oxidation states of transition metals	Grenier J; Pouchard M; Wattiaux A
16	11	● Obtention and study of new vitreous materials	Portier J; Tanguy B; Videau J
17	9	● Iron and aluminum oxyhalides ● Boron sulfide	Colin A; Portier J; Rouxel J

Table 3
Top 7 Hagenmuller delayed papers.

SB	Article title	Authors	Publication year	Journal	Citation number	B index
SB 1	Clathrate structure of silicon Na_8Si_{46} and Na_xSi_{136} (x < 11)	Kasper J.S., Hagenmuller P., Pouchard M., Cros C.	1965	Science	344	200.47
SB 2	New oxygenated bronzes of formula Na_xCoO_2 (x < =1) - cobalt-oxygen-sodium system	Fouassier C., Matejka G., Reau J.-M., Hagenmuller P.	1973	Journal of Solid State Chemistry	285	228.82
SB 3	Structural classification and properties of the layered oxides	Delmas C., Fouassier C., Hagenmuller P.	1980	Physica B+C	269	288.91
SB 4	Electrochemical behavior of the phases Na_xCoO_2	Braconnier J.-J., Delmas C., Fouassier C., Hagenmuller P.	1980	Materials Research Bulletin	109	223
SB 5	Electrochemical intercalation of sodium Na_xCoO_2 bronzes	Delmas C., Braconnier J.-J., Fouassier C., Hagenmuller P.	1981	Solid State Ionics	282	252.67
SB 6	Study of the Na_xCrO_2 and Na_xNiO_2 systems by electrochemical deintercalation	Braconnier J.J., Delmas C., Hagenmuller P.	1982	Materials Research Bulletin	77	264.32
SB 7	Electrochemical intercalation and deintercalation of Na_xMnO_2 bronzes	Mendiboure A., Delmas C., Hagenmuller P.	1985	Journal of Solid State Chemistry	128	254.74

2. Aim

The present study gives an overview of Hagenmuller's legacy by using scientometric analysis. We investigate his body of work in order to reveal his main topics of research as well as his main coworkers. We also study delayed recognition by looking for SBs. We then try to understand the reasons for this delay and the context of the renewed interest in those papers.

3. Methods

To collect the publications and citation data of Paul Hagenmuller, we used the Web of Science® database. A collection of 796 publications with their metadata were extracted from the database. Also using Web of Science®, 16,276 citations were harvested through 31 December 2015 and were exported in CSV format. Excel was used for statistics and calculations. The network of co-authors was created using VOSviewer, an open-source software for bibliometric mapping [19]. Also using VOSviewer, bibliographic coupling was applied to Hagenmuller's publications in order to identify the main topics of research that he pursued with his coworkers. In a bibliographic coupling network, the relatedness of papers is based on the number of references they share. After excluding the unconnected single nodes, the resulting bibliographic coupling network is clustered into sub-groups of related papers, with the minimum possible cluster size being set to 3. The clustering is done using VOSViewer's clustering technique [20,21]. The identified clusters of papers are then analyzed in order to identify the main topics of research pursued by Hagenmuller and coworkers with the help of expert interviews (see below last paragraph) and the literature. The co-word map describing Hagenmuller's overall scientific production was created using Gephi, an open-source software for network analysis [22]. The co-word map contains 202 keywords. Betweenness centrality was used to determine important nodes in the co-word network [23].

The H-index of Paul Hagenmuller was calculated from the beginning of his career until 2015. Let N_p be the number of papers a scientist published over n years, a scientist has index h if h of his or her N_p papers have at least h citations each and the other $(Np - h)$ papers have $\leq h$ citations each [24].

To identify SBs in a corpus of papers, we calculate the “Beauty coefficient” (B), a parameter-free index introduced by Ke et al. [17]. “B” quantifies the extent to which a paper could be considered a SB by adding up differentials between the citation curve of the publication and a reference line calculated between the year of publication and the year of maximum citations. Applying their criteria to their database, Ke et al. found that the top 1000 SBs in their database correspond to papers with B ≥ 317.93.

Using Ke et al.'s criteria [17], the “Beauty coefficient” B was calculated for all of Hagenmuller's papers. The papers were then ranked by decreasing values of B.

The quantitative analysis conducted to understand the phenomenon of delayed recognition in Hagenmuller's body of work was complemented by a historical and sociological analysis of the context of production and publication of the SBs and their subsequent awakening. Semi-structured interviews were conducted with some of his main coauthors and coworkers: Christian Cros, Claude Delmas, Michel Pouchard, and Jean Sénégas. Based on those interviews, the publication trends of the topics “silicon clathrate”, “fullerene”, “sodium cobaltate”, and “Na-ion batteries” were plotted using the following respective queries performed in the Web of Science® database: (i) "clathrate" AND ("Silicon" OR "Si"), (ii) "Fullerene", (iii) "sodium cobaltate" OR "Na_xCoO_2", (iv) "na ion battery" OR "sodium ion battery" OR "na ion batteries" OR "sodium ion batteries".

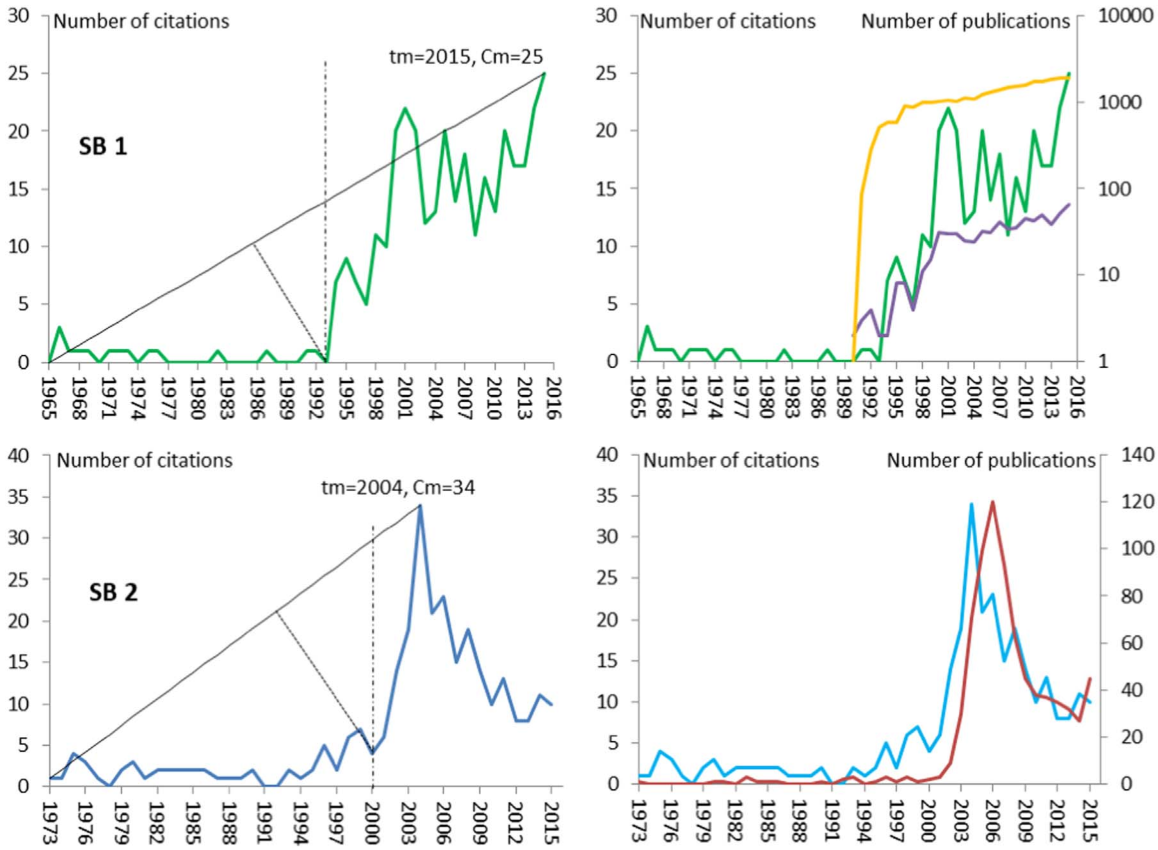

Fig. 4. Citation history of SB 1 and SB 2 and publication trends of silicon clathrates, fullerene, and sodium cobaltate. In color, yearly citations of SB 1 (*green*) and SB 2 (*blue*); distribution of publications on silicon clathrate *(purple)*; distribution of publications on fullerene (*orange*); distribution of publications on sodium cobaltate (*red*). *Black lines*, reference line l_t; *dotted lines*, distance d_t maximizing the awakening time; *vertical lines*, awakening time; c_m is the maximum number of citations; t_m is the year of the maximum number of citations c_m (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Results and discussion

4.1. Paul Hagenmuller's scientific production

Throughout a long scientific career of 54 years, Paul Hagenmuller published 796 papers. We can distinguish three periods in Hagenmuller's career:

The first period, from 1945 to 1956, marked his beginnings as a PhD student and as a research fellow at the CNRS. In this period, he published only 10 papers, mainly in French journals (Fig. 1).

The second period surrounded his appointment as an associate professor in Rennes from 1956 until 1961. During this period, he published 20 papers thanks to his greater autonomy and his capacity to recruit PhD students.

The third period spanned 41 years of research in Bordeaux (from 1961 to 2002). During this period, he was the author of 733 articles, 1 review, and some 42 other publications (conference proceedings, editorials, notes, etc.). All along his scientific career, he published in various sub-fields of chemistry (materials science, inorganic and nuclear chemistry, physical chemistry, etc.) in a wide variety of journals (Table 1). In particular, he was the fourth most published author in the *Journal of Solid State Chemistry* with a total of 110 papers.

Language-wise, he published mainly in French (Fig. 1; *blue columns*), English (Fig. 1; *orange columns*), and occasionally in other languages like German (Fig. 1; *black columns*). He transitioned slowly from publishing almost exclusively in French to privileging English for scholarly communication. 1981 was the year in which he started writing more than half of his yearly publications in English.

Hagenmuller collaborated with a large number of researchers through the years (Fig. 2), particularly his PhD students and fellow researchers in Bordeaux such as Michel Pouchard, Jean Ravez, Jean-Maurice Reau and Gérard Demazeau (150, 106, 92, and 78 co-publications respectively). But he also collaborated with other renowned international physicists and chemists including John B. Goodenough (The University of Texas at Austin, TX, USA) and John S. Kasper (General Electric, NY, USA).

Along with his coworkers, Paul Hagenmuller investigated a wide range of phenomena with a twofold strategy in mind: elaborating well defined materials with complex compositions or textures while also determining their atomic structure, measuring their physical properties, and accounting for their physical behavior in a comprehensive way [25]. This methodological strategy is clearly visible in the co-word network analysis (Fig. 3), where keywords such as “crystal structure” and “electrical properties” got the highest betweenness centrality values (8059.73 and 3488 respectively). The co-word network also shows the various areas investigated such as ionic conductivity, optics, ferroelectricity, as well as the various compounds studied (oxides, fluorides, cobalt, clathrates, intercalation compounds, ferroelectric materials, etc.). The bibliographic coupling network of Hagenmuller's publications contains 17 clusters of related papers. Table 2 provides some of the main topics of research as well as the top 3 co-authors in each cluster of the bibliographic coupling network.

Paul Hagenmuller's work was recognized in the academic community (1382 citations in 1980); by 2015, his work had been cited 16,276 times (Fig. 1; *dashed line*) and his H-index reached 58 in the same year (Fig. A.1 in Appendix A in the Supplementary Information). Particularly, the citation dynamics of Hagenmuller's work have witnessed a notable evolution since 2010 after being fairly stable for more than 20 years. Indeed, the average growth rate of Hagenmuller's citation curve went from −3.23% in the 2005–2010 period to 12% in

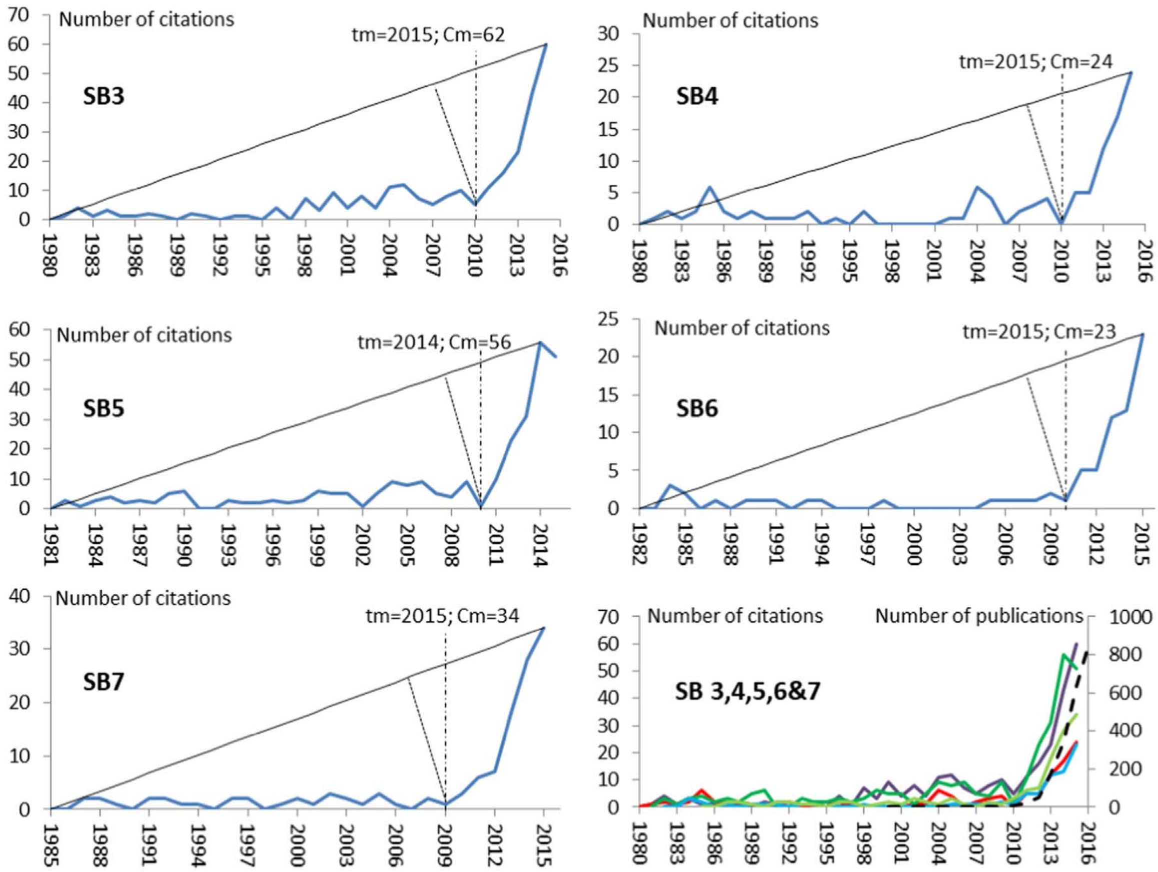

Fig. 5. Citation history of SB 3, SB 4, SB 5, SB 6, SB 7, their citation network, and sodium-ion battery publication trends. In color, yearly citations of SB 3 (*purple*); SB 4 (*red*); SB 5 (*dark green*); SB 6 (*blue*); SB 7 (*light green*); *black lines*, reference line l_t; *dotted lines*, distance d_t maximizing the awakening time; *vertical lines*, awakening time; *Black dashed line*, yearly publications on sodium-ion batteries per year (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the 2010–2015 period (Fig. 1; *black line*). Moreover, his H-index kept increasing every year even though his last paper was published in 2006 (Fig. A.1 in Appendix A in the Supplementary Information). This strengthens the hypothesis that some of his work was undergoing delayed recognition.

4.2. Investigating delayed recognition

The calculation of the “Beauty coefficient” for all of Hagenmuller's papers provides a ranking of these publications according to their respective B index value. For the purpose of this study only the top 7 papers are considered, which correspond to papers with $B > 200$ (Table 3). This means that those papers belong to the 1% of papers with most delayed recognition according to Ke et al. [17].

SB 1 [26] is a paper that belongs to cluster 12 (see Table 2). It was published in 1965 in *Science*. It made the link, for the first time, between the crystal structure of two cubic phases of the silicon-sodium system and clathrate type hydrates. SB 1 ‘slept’ for 28 years before it was awakened in 1993 (Fig. 4; *green curve*). The awakening of SB 1 coincides with a noticeable increase in publications on silicon clathrates (2 publications in 1993, 65 publications in 2015) (Fig. 4, *purple curve*). It is also contemporary to the emergence of fullerene research as a hot topic in the 1990s (1 publication in 1990, 1906 publications in 2015) (Fig. 4, *orange curve*). Indeed, the discovery of fullerenes in the late 1980s [27] and the awarding of the Nobel Prize in Chemistry in 1996 to Robert F. Curl Jr, Harold W. Kroto, and Richard E. Smalley drew attention to silicon clathrates, and awakened SB 1.

SB 2 [28] is a paper that belongs to cluster 8 (see Table 2). It was published in 1973, and was written in French. Fouassier et al. reported the isolation of four bronze type phases with the formula Na_xCoO_2 (x $<=1$) by investigating the sodium-cobalt-oxygen system. The paper ‘slept’ for 27 years before it was awakened in 2000 (Fig. 4; *blue curve*). The citation history of the paper can be associated with an increase in publications on sodium cobaltate (2 publications in 2000, 120 publications in 2006) (Fig. 4, *red curve*). In fact, it was the discovery of new properties, namely the large thermoelectric power of layered sodium cobalt oxides [29,30], as well as their superconductivity [31] that renewed interest in this oxide system, and subsequently awakened the SB.

SB 3 [32], 4 [33], 5 [34], 6 [35], and 7 [36] are all papers that belong to cluster 8 as well (Table 2). They were published between 1980 and 1985. SBs 4 and 6 were written in French. In addition to Paul Hagenmuller, the main investigators were Claude Delmas and Claude Fouassier. They mainly investigate layered oxides (properties, structural classification), as well as electrochemical intercalation and deintercalation processes of various compounds. The sleeping period of those papers ranges between 24 and 30 years. The awakening of those SBs was almost simultaneous (Fig. 5), with SB 3, 4, 5 & 6 awakening in 2010 and SB 7 awakening in 2009. The awakening seems to match an increase in Na-ion battery research since 2010, with the number of publications rising from 6 in 2010 to 649 in 2015 (Fig. 5; *black dashed curve*). The collective awakening around 2010 can be explained by the fact that sodium-based batteries became regarded as alternatives to lithium batteries. In the 1970s and 1980s, both Na-ion and Li-ion batteries were investigated, but the higher energy density of Li-ion cells made them more applicable to small, portable electronic devices, concentrating research efforts on Li-ion batteries from that time [37]. Despite the maturity of lithium-ion battery technology, questions remained about the safety, lifetime, poor low-temperature performance, and cost of lithium batteries [38]. On the other hand, sodium is widely available in the earth's crust and sea, and is the second lightest alkali after lithium [39].

Since Van Raan [16], it is common to refer to the concept of the “prince” in order to characterize the papers that awakened a SB. A discussion of the relevant princes of Hagenmuller's seven SBs is left to a future work.

5. Conclusions

The scientometric analysis of Paul Hagenmuller's body of work enabled us to reveal the richness and diversity of his contributions to the development of solid-state chemistry. During his long and productive career, Hagenmuller played a key part in the creation and development of an independent and successful solid state chemistry adjacent-field to materials research.

Although Hagenmuller was a recognized scientist, both nationally and internationally, that did not prevent part of his work from getting attention from the scientific community at a later time. This delay can mainly be explained by the lack of immediate relevance of those findings to the existing state of knowledge at the time of publication. This is not an unusual phenomenon in curiosity-driven basic research. However, other factors may explain the lack of diffusion of those publications, such as language choice. In fact, three out the seven SBs reported in this study were published in French (SBs 2, 4 and 6). Therefore, it could be hypothesized that a significant proportion of the solid-state chemistry community did not have access to the content of those papers due to linguistic limitations. A quantitative investigation of the issue of language use in research has been previously investigated in scientometrics [40,41], but never as a probable cause of delayed recognition. Such investigation is left for future work, though it is relevant to note that this hypothesis was dismissed by most interviewed experts.

At the individual level, although SBs are far from being exceptional phenomena in science, it is very striking that a single scientist co-authored seven of them. The high number of SBs in Hagenmuller's work originates from the fact that SB 2, SB 4, SB 5, and SB 6 deal with the same type of phases and/or compositions, and that SB 7 is a related phase system (Mn is substituted to Co). It could also be hypothesized that Hagenmuller's research strategy and his focus on identifying and creating new materials favored the occurrence of SBs within his publications. In addition, newly identified compounds do not necessarily find industrial applications right away as exemplified by the sodium cobaltate materials. A long time could go by before new industrial applications appear.

At the field level, the occurrence of SBs varies considerably, and physics and chemistry are the fields that produce most SBs [17]. It would be of interest, then, to quantify the occurrence of SBs in subfields of chemistry such as materials science or solid-state chemistry. Also, the occurrence of SBs raises questions about the relevance of short-term citation-based metrics for the evaluation of scientific impact.

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Declarations of interest

None.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jssc.2018.02.003.

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