



Review

Systematic Review of Lithium-Ion Battery Recycling Literature Using ProKnow-C and *Methodi Ordinatio*

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Abstract: Recycling lithium-ion batteries (LIBs) plays an important role in environmental preservation since it prevents heavy metals from polluting the soil and underground water through the recovering of valuable metals. The interest in LIB recycling has grown in recent years due to the environmental and economic gains which can be seen by increasing number of articles and publications. This review uses two methodologies: ProKnow-C and *Methodi Ordinatio* to create a bibliographic portfolio (BP) that defines the state-of-the-start literature in LIB recycling. This review is vital because it proposes a database of a finite number of publications of relevant authors and articles to service new research on the LIB recycling theme. The research started off with 2515 articles related to the search query which were later filtered and treated to be systematically analyzed. After filtering, 591 articles were left in the filtered raw article database (FRA-database). The efficiency and parameters of ProKnow-C and *Methodi Ordinatio* were counter-compared forming two databases. These databases were analyzed systematically and it was found that in the initial stages there were no differences between them. Nevertheless, in the final phases, a difference in the ranking was established when compiling the final BP of the 23 best ranked articles and authors. By using ProKnow-C and *Methodi Ordinatio*, this review sets out to establish a concise BP of paramount importance to the LIB recycling theme.

Keywords: lithium-ion battery; valuable metals; recycling; ProKnow-C; *Methodi Ordinatio*



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1. Introduction

Since its debut in 1991, lithium-ion batteries have grown their outreach and have gone from being an option amongst other batteries, to being the leader and standard for electronics, such as cameras, notebooks, cell phones, and other gadgets [1–5]. The introduction of lithium-ion rechargeable batteries (LIBs) by Sony in the 1990s established a new frontier for electronics usage, enabling engineers to push the boundaries and set new parameters for upcoming technology use [6–9]. In the last decades, global research efforts caused LIBs to stand out due to their innate characteristics: storage capability, rechargeability, highly efficient charging, weight (lightweight), constant power, long shelf life, more usable capacity than lead-acid equivalent, amidst others [10–12]. LIB production and market demand grew rapidly throughout the years and nowadays, the two major segments for LIB usage can be categorized into automotive and nonautomotive [3,7–9,13].

In both segments, the demand for LIBs has grown exponentially and has projected growth. The battery-run automotive segment was always promising, but due to its low mileage, long charging time, and high cost compared to fuel-driven automobiles, electronic vehicles (EV) were seen with skepticism. Today, in virtue of great advances made in the EV sector, the demand and projected growth estimate this to be a \$67.2 billion market by 2025 [14]. This means that by 2030 around 140 million EVs are predicted to cruise the roads [10,15]. Since EVs need automotive batteries to run, LIBs have predominated the

sector due to their aforementioned qualities. Another great quality of EV-LIB is that they have a second-life usage. In other words, the LIB from EV which is no longer suitable for EV application is inserted into another sector, proving added value to the batteries. Battery reuse enhances the circular economy and is a path towards sustainability [3,7,16]. Nonautomotive LIB usage is very ample and can be found in smartphones, electronic gadgets, medical equipment, power tools, solar power storage, emergency power backups, alarm systems, and others. Since LIBs have shown the best result in these applications, they should continue to hold the market for some time longer. Therefore, projections that the LIB market will reach \$94.4 billion by 2025 indicate the economic and social value that LIBs have to offer [17].

Despite these qualities, it is necessary to understand that if LIBs in end-of-life (EoL) are not properly disposed of, they can become an environmental and health hazard. Therefore, after fulfilling their primary and secondary usage, LIBs need to be recycled or recovered properly. In many countries, especially in developing nations, LIBs are sent to landfills and treated as common solid waste. This can cause harmful toxic substances to end up in groundwater. Besides being an environmental contaminant, these so-called “contaminants” are in fact valuable metals that can be reinserted into the economy. One of the drawbacks of using lithium-based batteries is that the extraction of lithium is extremely polluting. Another disadvantage is that the price of lithium, cobalt, nickel and other valuable metals has gone up due to LIB's growing demand. By recycling LIBs, these valuable metals never become environmental contaminants, and they can have, yet again, commercial value. Another interesting aspect to consider is that mining will no longer be the sole source for metal extraction. Hence, LIB manufacturers will have another option for acquiring these valuable metals creating a competing market, and possibly lowering the cost of these valuable metals based on the Law of Supply and Demand [13].

This article approaches the LIB recycling theme with an in-depth review to find the state-of-the-art using two bibliometric methods comparing the results between them. The methodologies Knowledge Development Process-Constructivism (ProKnow-C) and the *Methodi Ordinatio* have been used to support systematic reviews. Both bibliometric methods have the same number of phases (9 total), despite not being totally equivalent, which can be summed up into two main phases: preliminary investigation (identifies the raw database) and portfolio filtering (which selects aligned and relevant publications). There is a lacuna in the literature when it comes to comparisons between the methods and their results [18–22]. This paper applies both methods simultaneously to select relevant publications and compare the results between them.

2. Materials and Methods

The many works done by distinct researchers give an ample and overwhelming database for review articles such as this one. Therefore, one of the most important steps when writing a review article is to find articles and discussions which pertain directly to the objective at hand. These findings make up the bibliographic portfolio (BP).

This article presents a systematic literature analysis using the ProKnow-C methodology, which is widely recognized and has been used in reviews of scientific articles. In the ProKnow-C methodology, the most relevant publications, authors, keywords, and journals are identified according to a specific research theme.

The ProKnow-C method is made up of three main phases [18–20]: (A) Selection of the bibliographic portfolio (BP); (B) bibliometric analysis of the BP; and (C) systemic analysis of the BP.

2.1. Methodology: ProKnow-C

ProKnow-C consists of a series of procedures that aid the researcher through the filtering and selection of articles that are relevant to the research theme. The method is set up into two phases: (1) Sets up an unfiltered raw article bank (URA-database); and (2) a selection process of the collected database.

These steps generate the bibliographic portfolio (BP) selection phase. Based on the research theme, the following axis, keywords, and search engine was established.

2.1.1. Phase 1: Raw Article Bank–BP

The first phase takes into account the research theme, and tries to answer the question, “What is the state-of-the-art for lithium-ion battery recycling processes?”. To do so, a systematic approach was put into place, and the following steps were established for the first phase: (1) Defining Keywords; (2) query and research, and (3) results and filtering.

The conclusion of these steps, set up a filtered raw article bank (FRA-database) also defined as filtered raw bibliographic portfolio which later underwent phase 2: Bibliometric analysis of the BP and Phase 3: Systematic analysis of the BP.

Step 1: Defining Keywords

The keywords take into account an overview of the research parameters, which can be overlapping and/or isolated, nevertheless linked in some way to the chosen theme as seen in Figure 1.

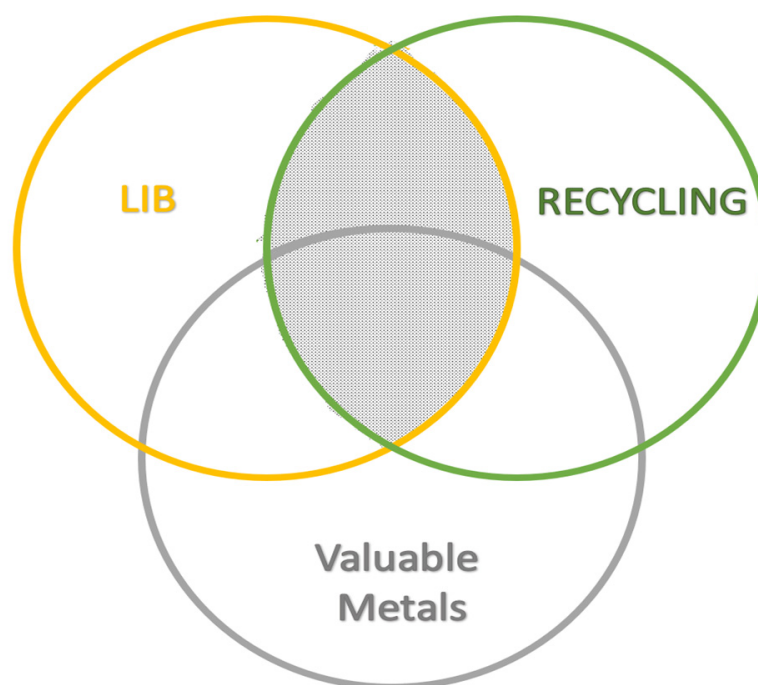


Figure 1. Research Parameters.

Keywords are extracted from the research axes or research parameters. Therefore, in this initial step, the range of topics is limited based on the chosen axes. These were defined as the research parameters: (A) Lithium-ion batteries; (B) recycling (lithium-ion batteries); and (C) valuable metals.

Based on these three axes, keywords and their synonyms were chosen for each one. For the first axis, lithium-ion batteries, the following keywords were used: lithium ion batteries, LIBs, LCO (lithium cobalt oxide), LPF (lithium iron phosphate), LMO (lithium manganese oxide), NCA (lithium nickel cobalt aluminum oxide), NCM (lithium nickel manganese cobalt oxide), LTO (lithium titanate). For the second axis, recycling, the following keywords were used: recycling, recovery, reuse, recondition. For the last axis, valuable metals: precious metals, rare metals, valuable metals, cobalt, copper, lithium, nickel, manganese, aluminum were used as keywords.

Step 2: Query and Search

The keywords were linked using the logic expressions (and) (or) forming 288 possible combinations, which defines the scope and specificity of the research. Using these keywords and their synonyms, together with the use of wildcards, such as \$ and * alongside the logic expressions: and/or—the query shown in Figure 2 was formed. This query was fed into the search engines: SCOPUS and Web of Science (WoS).

(lithium ion batter* or LIB* or Lithium Cobalt Oxide or Lithium Iron Phosphate or Lithium Manganese Oxide or Lithium Nickel Cobalt Aluminum Oxide or Lithium Nickel Manganese Cobalt Oxide or Lithium Titanate or LCO or LPF or LMO or NCA or NCM or LTO and Recycl* or recover* or reuse or recondition and Valuable metal* or preciou* metal* or rare metal* or valuable metal* or Cobalt or Copper or Lithium or Nickel or Manganese or Aluminum)

Figure 2. Search Query. Wildcard asterisk (*) is a substitute that ranges from 0 to infinite characters.

Step 3: Results and Filtering

The search results from 27 November 2021 generated 78 document results for SCOPUS and over 4 million (4,222,313) results for WoS. These results compile the unfiltered raw article database (URA-database). After establishing the URA-database, filters were placed in motion to limit the number of search results from the predefined query.

Two types of filters were used: (1) Date filtering was established after 1991—the year in which the commercial use of LIBs came into play; and (2) specific types of papers were chosen: articles and reviews. The new search results with both filters in place generated 78 results for SCOPUS and 2515 results for WoS, which make up the filtered raw article database (FRA-Database).

Some articles were chosen at random to prove the efficiency of the keywords and to see if new keywords were to be inserted. It was found that the search parameters were adequate, and no new keywords were necessary. The description of Phase 1 is shown in Figure 3.

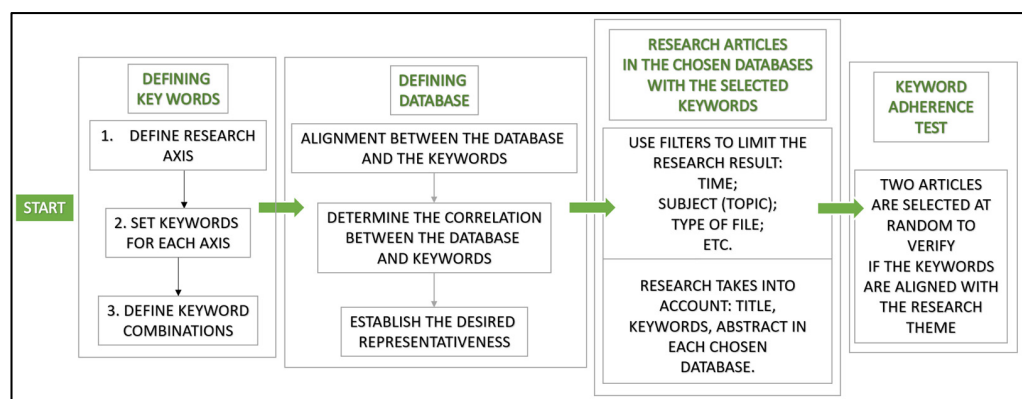


Figure 3. ProKnow-C method adapted from [18]. Description of Phase 1—Setting up an FRA-database.

2.1.2. Phase 2: Selection Process

The second phase starts with the exclusion of duplicate articles from the FRA-database. This is necessary especially when multiple search engines are used. In this review, since there were results from two distinct search engines (SCOPUS and WoS), it was necessary to eliminate duplicates. After doing so, there were a total of 2473 articles and review articles left in the database (out of the initial 2515).

The subsequent step is an overlook at the article title comparing it to the review theme. If the article's title is relevant and pertaining, the article remains in the database; otherwise, it is excluded. The result from this step left 591 articles in the database. The following step is to verify the scientific recognition of the articles left in the database. To do so, the number

of citations each article has is interposed with an arbitrary value selected by the study. By doing so, the articles with a relevant number of citations or newly published articles (2017 on) were taken into consideration for this review article. This step was performed using the *Methodi Ordinatio* and ProKnow-C and later the results were compared.

Methodi Ordinatio

After the title alignment with the research theme, the articles were analyzed based on the *Methodi Ordinatio*. To do so, some parameters had to be established as shown in Figure 4. Parameter 1: Alfa—is an arbitrary value between 1 and 10, where 1 refers to older published work and 10 to recent articles. Parameter 2: Search Year—it is the year in which the research took place, in this case, 2021.

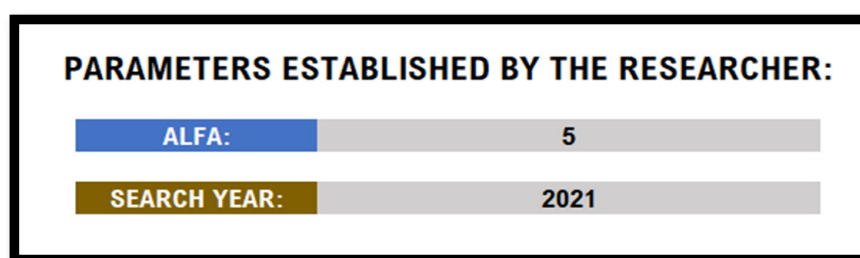


Figure 4. *Methodi Ordinatio* Parameters.

Using the parameters mentioned, as well as the Journal Citation Reports (JCR) impact factor and number of citations (Equation (1)), a value was assigned to each article, making it possible to rank the articles. The Pareto Principle was applied to exclude publications whose value was below the threshold.

The ranking number was established using the following equation:

$$Ordinatio = JCR + Citations + \alpha (10 - \alpha \cdot Age) \quad (1)$$

where *JCR* is the Journal Citation Report number for each article based on the Journal Impact Factor of each publication; *Citations* takes into consideration the number of times the article was cited; α (alpha) is a value between 1 and 10 which takes into consideration the newer and older articles, respectively; and *Age* refers to the age of the article in reference to the search year (2021).

By performing and assigning ranking numbers to each article based on their *Ordinatio* number, a ranked list of 588 articles was formed in the database, making it possible to perform a systematic analysis of the BP which can be seen in the Results and Discussion section.

ProKnow-C

To compare results between the *Methodi Ordinatio* and ProKnow-C, a ranked database was also formed using the ProKnow-C method. In this method, the number of citations is the key vector, since the citation percentage is calculated for each article.

$$Citation \% = \frac{Article\ Citation}{\sum Database\ Citations} \quad (2)$$

Later, the accumulated percentage of *citations* was calculated and the Pareto Principle was also applied. These articles formed the initial ProKnow-C database.

The next step was to observe the articles which did not make the cut. These were analyzed based on two factors: (1) Recent articles and/or (2) Common Authorship. The first-factor analysis articles that even though did not make the cut in the accumulated percentage citation step, could be included in the database if they were considered to be recent or new articles. In this study, recent articles were defined as published from 2017 on. The second factor takes into account the authors and coauthors. In this step, all the authors

left out during the Pareto and Recent Article phase were cross-examined with the ones who made the cut. By doing so, authors who appeared in the first two steps can have their uncited, less cited or “old” articles also added to the database. The overview description of Phase 2 is shown in Figure 5.

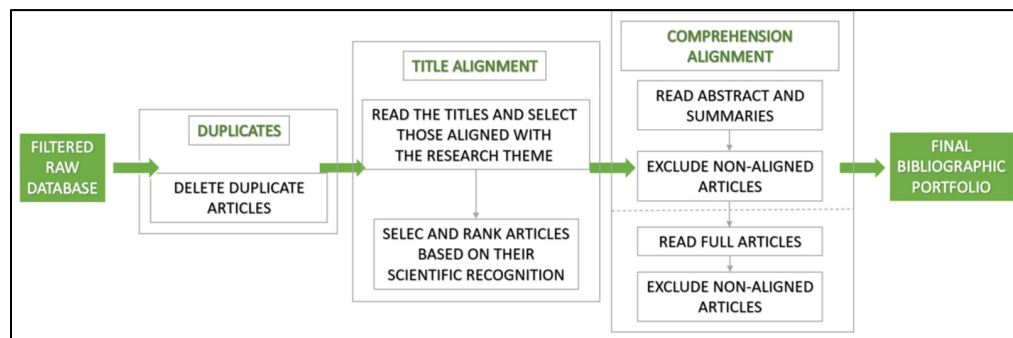


Figure 5. ProKnow-C method adapted from [18]. Description of Phase 2—Generating final bibliographic portfolio.

The last step is twofold; first the availability of the full articles are determined. Those which are not available are then excluded from the BP. To finalize the last step, it is necessary to determine the alignment of the articles kept in the final database. To do so, the articles must be read and non-aligned articles must be excluded. Based on the results from both analyses, *Methodi Ordinatio* and ProKnow-C, the results for the bibliographic portfolio were obtained and analyzed as described in the Results and Discussions section.

3. Results and Discussion

In the initial exploration, search engines SCOPUS and WoS retrieved more than 4 million relevant publications due to the specificity of the search query. To limit these results, filters (Year: after 1991 and Document Type: Reviews and Articles) were applied. By doing so, the search result came down to 2515 articles. Since two types of search engine were used, it was necessary to exclude duplicates, leaving the filtered raw article database (FRA-database) with a total of 2437 articles. The titles of these articles were read and if they were aligned with the theme, they were kept in the database. Otherwise, they were removed. The final result left the FRA-database with 591 articles.

These articles from the FRA-database were analyzed using two methodologies: (1) *Methodi Ordinatio*, and (2) ProKnow-C. Article similarity to the research theme, JCR, number of citations, age, and common authorship were analyzed. After establishing the bibliographic portfolio (BP), a systematic analysis was performed to best understand the search results.

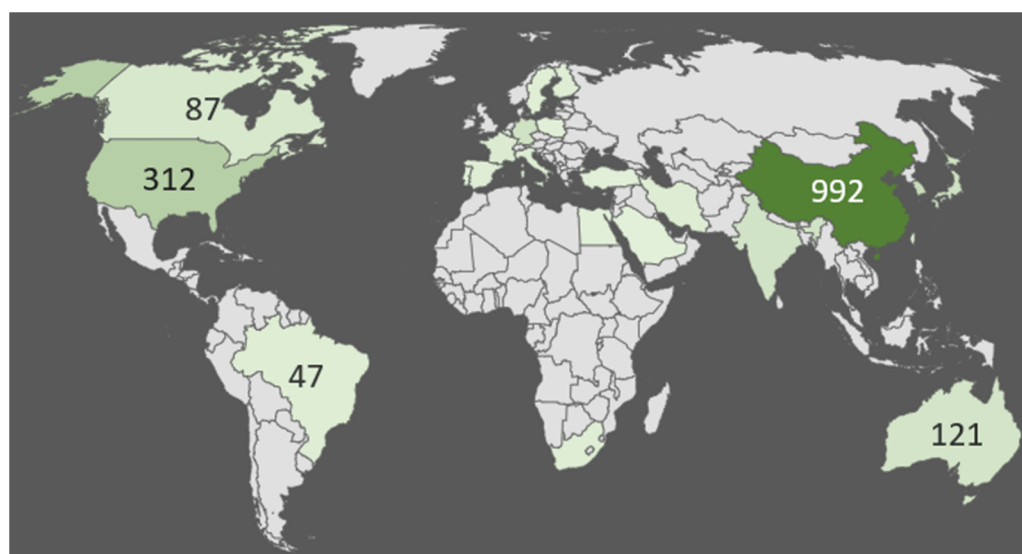
3.1. Countries

The number of articles that make up the BP is 591. Out of these, the countries with the greatest number of publications were the People’s Republic of China (40.6%), the USA (12.6%), South Korea (7.9%), Japan (6.2%), and Germany (5.8%). Table 1 shows a list of the countries which have more than 1% of publications in respect to LIB recycling out of the total BP and Figure 6 shows a demographic distribution of the countries in Table 1.

It is clear that China is by far the most active in article publication regarding LIB recycling with 27.6% more publications than the USA, which is the second-largest researcher. China’s leadership in LIB recycling publications can be attributed to the fact that China is the largest consumer of electronics and therefore produces the highest number of spent LIBs [23–28]. To manage such waste, China has issued specific legislation to deal with battery waste and to promote valuable metal recycling, as mentioned by [29].

Table 1. Countries with at least 1% publication on LIB recycling.

Countries	Number of Publications	Percentage (%)
China	992	40.2
USA	312	12.6
South Korea	195	7.9
Japan	153	6.2
Germany	142	5.8
India	132	5.3
Australia	121	4.9
Canada	87	3.5
England	79	3.2
France	67	2.7
Italy	55	2.2
Iran	52	2.1
Finland	48	1.9
South Africa	48	1.9
Brazil	47	1.9
Spain	47	1.9
Sweden	40	1.6
Singapore	33	1.3
Taiwan	33	1.3
Belgium	29	1.2
Turkey	25	1.0

**Figure 6.** Publication per year–demographic distribution with the number of publications in the LIB recycling field.

3.2. Research Group Affiliation

When analyzing the research group affiliation of each country, out of the 25 highest ranked research groups, China has 12 representatives, the USA has 4, India and Germany 2 each, and Australia, France, Finland Korea, and Japan have one research group each as seen in Figure 7. The research group with the greatest number of publications is the Chinese Academy of Sciences with a total of 137 publications out of the 591 articles from the BP.

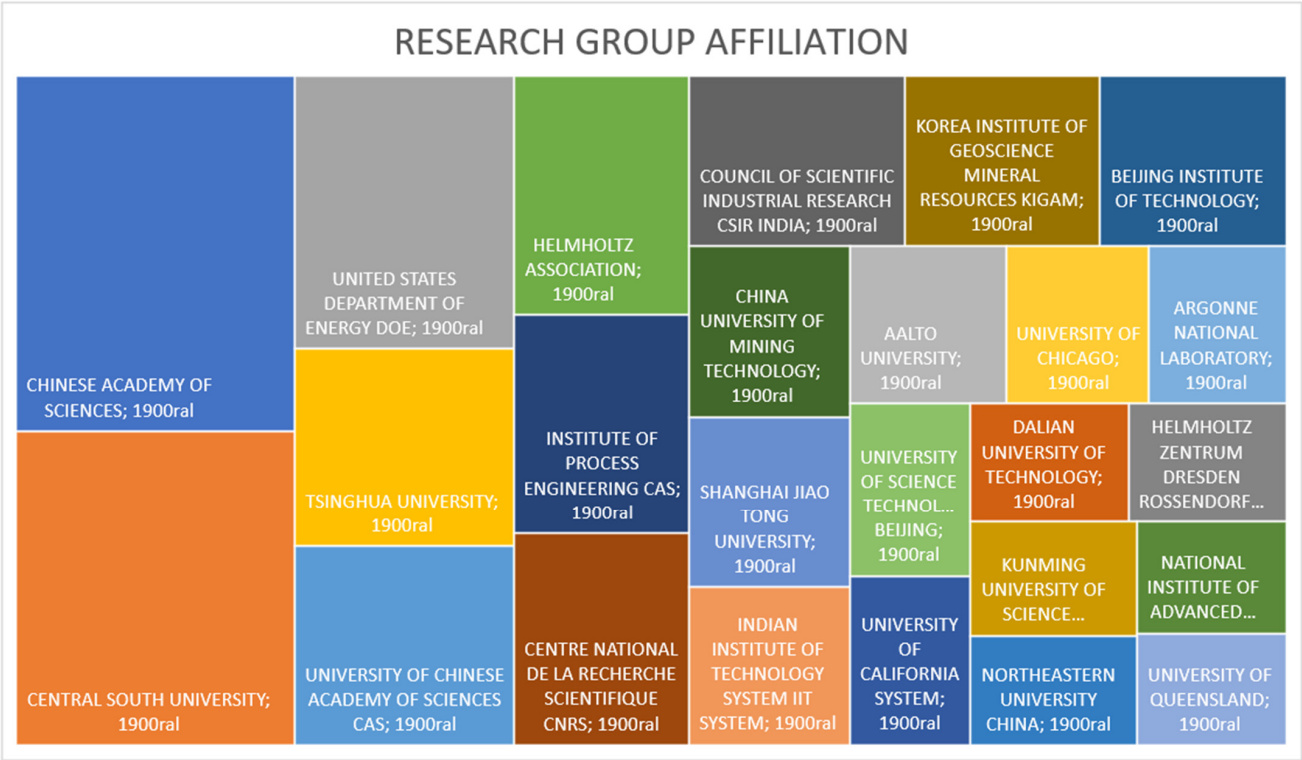


Figure 7. LIB research group affiliation—top 25 groups with most publications in the LIB research field.

3.3. Publications per Year

The interest in LIB recycling and research has grown exponentially in the last few years due to technological advances for portable devices and also EVs [16,24,25,30]. During the first two decades of the LIB (1991 up to 2011), the number of publications regarding LIB recycling had not reached 50 yearly publications, meaning that there was little research or not enough reasons to pursue the research theme. After 2012, the number of published articles grew somewhat exponentially, reaching its peak of 438 yearly publications in 2021, as shown in Figure 8. The average yearly publication growth is 19% taking into consideration the years 2000 to 2021.

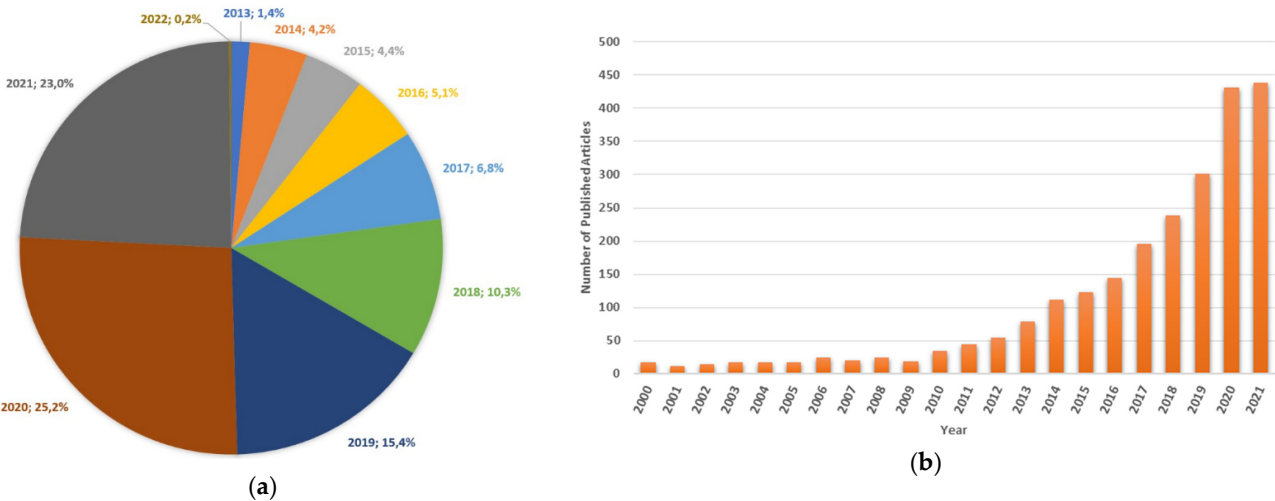


Figure 8. Yearly publications on LIB recycling: (a) graphical and (b) table format.

3.4. Publications per journal

Another analysis performed was in regards to the journal where the articles were published. The top 10 journals with the greatest number of LIB recycling articles are listed in Figure 9.

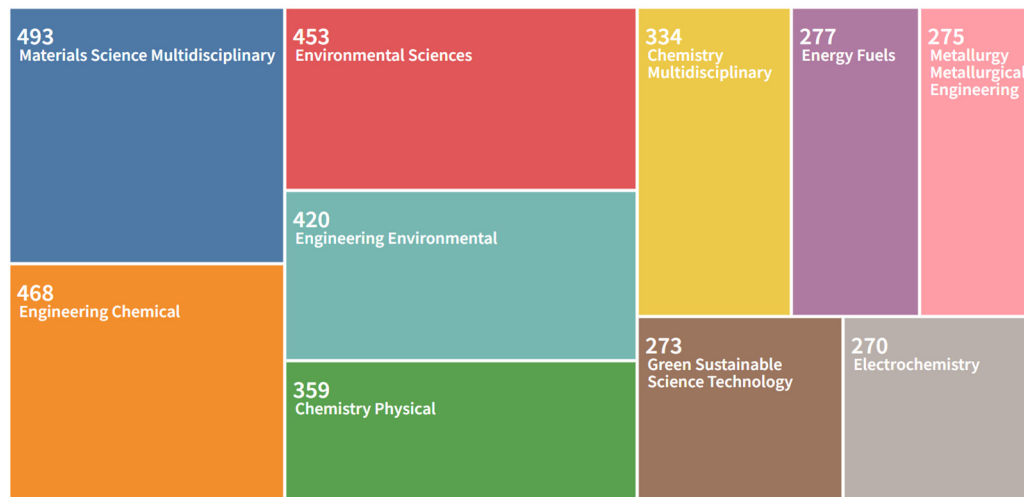


Figure 9. Top 10 journals with published LIB recycling articles.

Based on the type of journal and research area it is possible to establish the areas affected by LIB recycling, such as materials science, whose objective is to find new or alternative materials to be used as a cathodic material, for example [31–33]. LIB recycling is fundamental for the Chemical Engineering field, as it focuses on the enhancement of new materials and processes, but also the use and reuse of waste material. From spent batteries, waste materials are valuable materials that can be used in new material compositions.

Not recycling LIBs is by far the worst option, since it wastes valuable metals which have financial value and are just buried away. The other perspective is the environmental outlook, which ensures that improper discarding leads to environmental problems, such as soil contamination, pollution, and health hazards to the population [5,34].

3.5. Keywords

The areas of LIB recycling are very diverse since they range from pre-treatment methods to cathode relithiation [35,36]. Therefore, another interesting analysis was to verify the keywords used in the articles selected by the *Ordinatio* and ProKnow-C methodology. Table 2 shows the 10 most used keywords and their occurrence.

Table 2. Most used keywords in LIB recycling published work.

Keywords	Occurrences	% of Occurrence
Cobalt	259	11%
Lithium-ion battery	245	10%
Recovery	229	9%
Valuable Metals	220	9%
Lithium	204	8%
Separation	169	7%
Recycling	151	6%
Cathode Materials	94	4%
Hydrometallurgical Process	88	4%
Acid	82	3%
Sum	1741	71%

From the 591 articles selected, the sum of the keyword occurrences from Table 2 adds up to 1741 occurrences for these keywords. This means that these top 10 keywords make up more than 70% of all keyword occurrences in the FRA-database which takes into account all relevant publications about LIB recycling.

This keyword occurrence result is extremely useful for new researchers, since it allows them a shortcut to the most relevant work in the LIB recycling field. It is also interesting to observe that the number one keyword use goes to cobalt. This can be explained due to the fact that the most valuable metal, in financial terms, in a spent battery is cobalt [37–39]. Other factors, such as the increased demand, limited and depleting reserves, and political-social factors hinder cobalt availability [40–42]. When comparing prices, it can be seen that the value of cobalt has been on the rise since 2018, when cobalt prices on the London Metals Exchange gradually fell from about 40,000 to 23,000 USD per ton between 2010 and late 2016. After that, the cobalt price rose again, reaching its peak at the beginning of 2018, at more than 90,000 USD per ton. The price dropped again reaching its baseline in mid-2019. Now the price has been on the rise again and is around 70,000 UDS per ton [13,43,44].

3.6. Number of Published Articles

To conclude the systematic analysis of the FRA-database, the authors and co-authors were analyzed. The FRA-database, or BP, which is composed of 591 articles, is made up of 2081 authors. Out of these, 2007 authors have less than 5 articles published in the LIB recycling field and only 13 have more than 10 articles published as shown in Table 3.

Table 3. Number of authors in respect to the number of published articles.

Number of Published Articles	Authors	Percentage (%)
Less than 5	2007	96%
5 to 10	62	3%
More than 10	12	1%
Total Number of Authors	2081	100%

The authors with the least number of publications amount to 1543. These authors only have one publication in the selected BP. Therefore, they were later analyzed to see their relevance and contribution to the LIB recycling theme.

The authors with the most publications in our database are shown in Figure 10: Li Li; Renjie Chen and Wu Feng all from Beijing Key Laboratory of Environmental Science and Engineering, School of Materials Science and Engineering, Beijing Institute of Technology, Beijing, China.

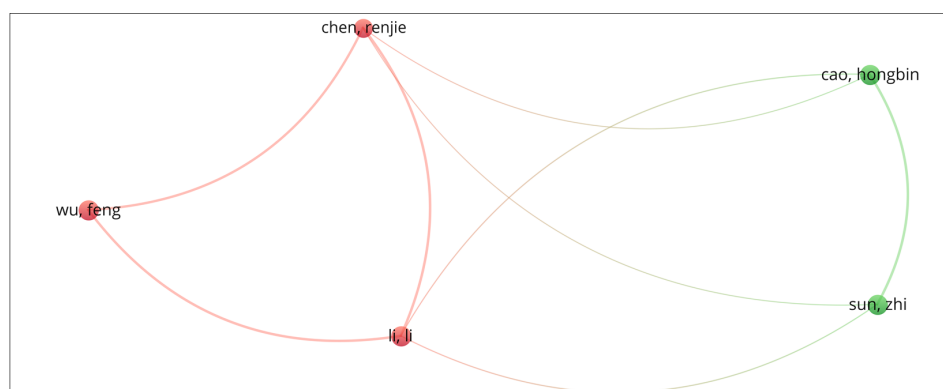


Figure 10. Authors with the greatest number of published articles in LIB recycling bibliographic portfolio—VOS Viewer Network Visualization.

Figure 10 shows two different clusters defined by the color red and green, which represent authors with more than 20 publications. These clusters have lines that indicate

links between the different clusters, meaning that there has been collaboration between authors. The three most published authors belong to the same green cluster and the distance between these authors in the visualization indicates the relatedness in terms of co-citation links. In general, the closer two journals are located to each other, the stronger their relatedness. The strongest co-citation links between journals are also represented by lines.

Another analysis performed on the highest-ranked authors in the BP was in regards to publication year, which can be seen in the Overlayer View in Figure 11.

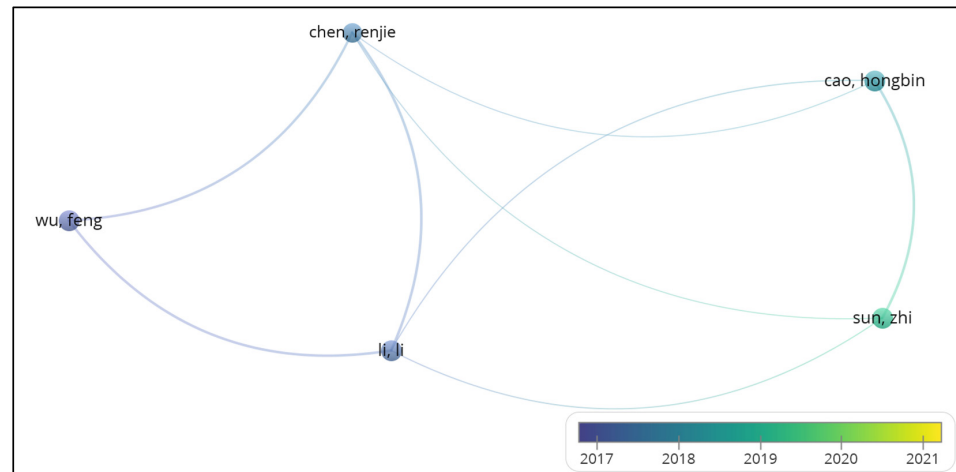


Figure 11. Authors with the greatest number of published articles in the LIB recycling bibliographic portfolio—VOS Viewer Overlay Visualization.

The overlay visualization is similar to the network visualization, except it is colored differently. In the network visualization, items are colored by different clusters, whereas in the overlay, visualization items can be colored based on the user-defined scores. In this representation, the color display considers the year of publication, ranging from 2017 to 2021. It can be seen that Wu, Feng; Li, Li; and Cheng, Renjie have older publications, while Cao, Hongbin, and Sun, Zhi have more recent articles. Nevertheless, none of these authors have articles published in 2021 that have made it into the LIB recycling BP.

Despite being an important factor, the year is not the most prominent parameter at this point. Therefore, these authors were also compared in regards to the number of citations for these selected articles, as seen in Figure 12.

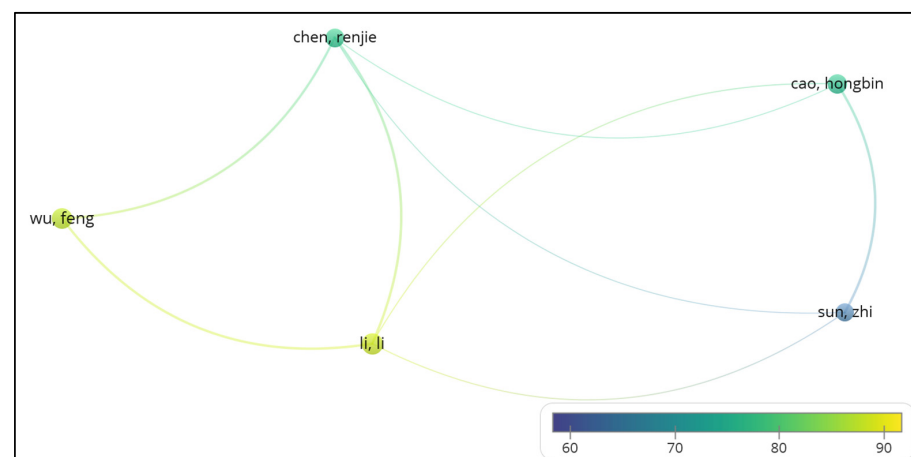


Figure 12. Authors with the greatest number of published articles in the LIB recycling bibliographic portfolio—VOS Viewer Overlay Visualization.

The number of citations ranges from 60 to 90. Authors with more yellowish colors are the ones who have more citations. This overlay visualization shows the average number of citations received by an author in which a keyword or term occurs. A simple correlation between Figures 11 and 12 proposes that older articles tend to show a greater number of citations. Nevertheless, this cannot be assumed to be true for all cases.

Continuing our primal analysis based on authorship, the 20 most published authors in the LIB recycling field can be seen in Figure 13. The initial observation only takes into account the number of total published articles throughout the years (2017–2021).

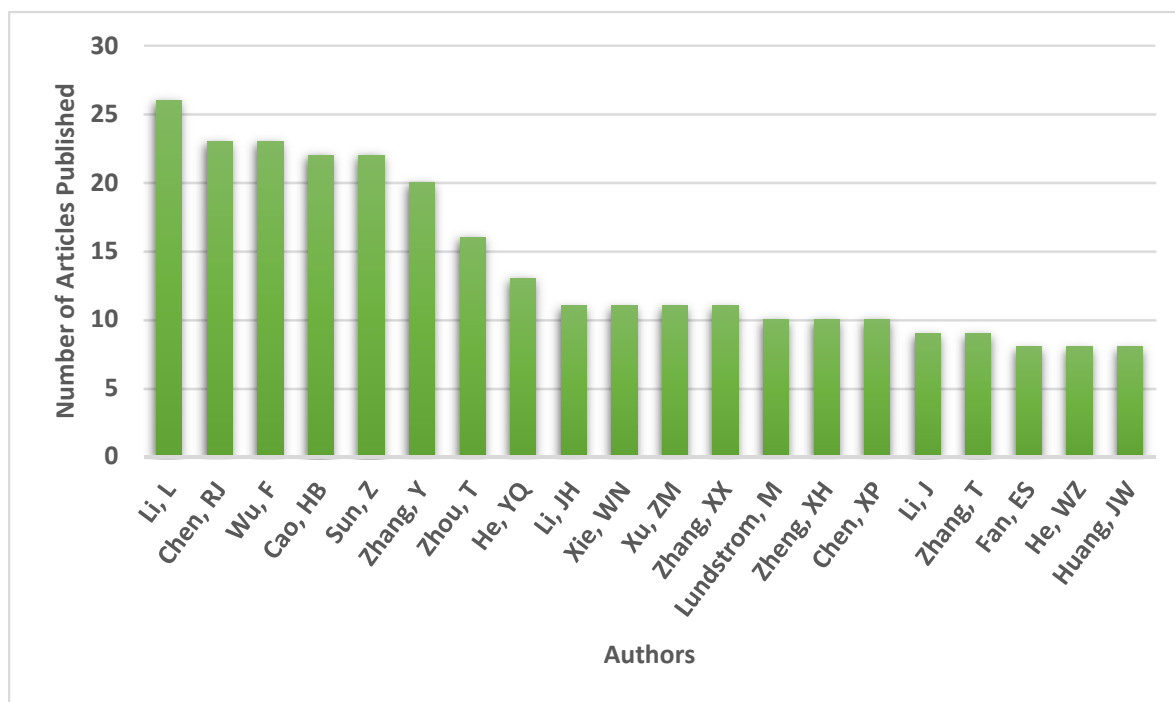


Figure 13. The 20 most published authors in the selected database.

As shown in Figures 10–12, the most published authors can also be seen in Figure 13 alongside others whose total number of publications ranges from 8 to 22 publications. It is worth noting that these are not all total distinct publications, for in many cases, these authors are coauthors in the same articles, as is the case with Li, Chen, and Wu.

3.7. Author Citation

The analysis of the remaining authors had to be done in regards to the number of citations, as most authors from the BP have fewer than five publications (96% as shown in Table 3). The number of times an author is cited is shown in Figure 14, along with its link to other authors.

In Figure 14 it can be seen that there are 4 distinct clusters which are based on the citation linkage between the authors. The clusters are composed of red: 53 authors; green: 21 authors; blue: 12 authors; and yellow: 11 authors. The greater the size of the label and circle, the more times the author has been cited. The lines link the author with those who have cited him/her. To exemplify, the author with the greatest number of total citations (1915 times) is shown in Figure 15.

Li, Li has been cited over 1900 times for all her work related to LIB recycling. Other authors like Wu, Feng; Cao, Hongbin Chen, Renjie have more than 1500 citations for all their published work in the selected BP. Other authors with more than 500 citations for all their published work can be seen in detail in Figure 16.

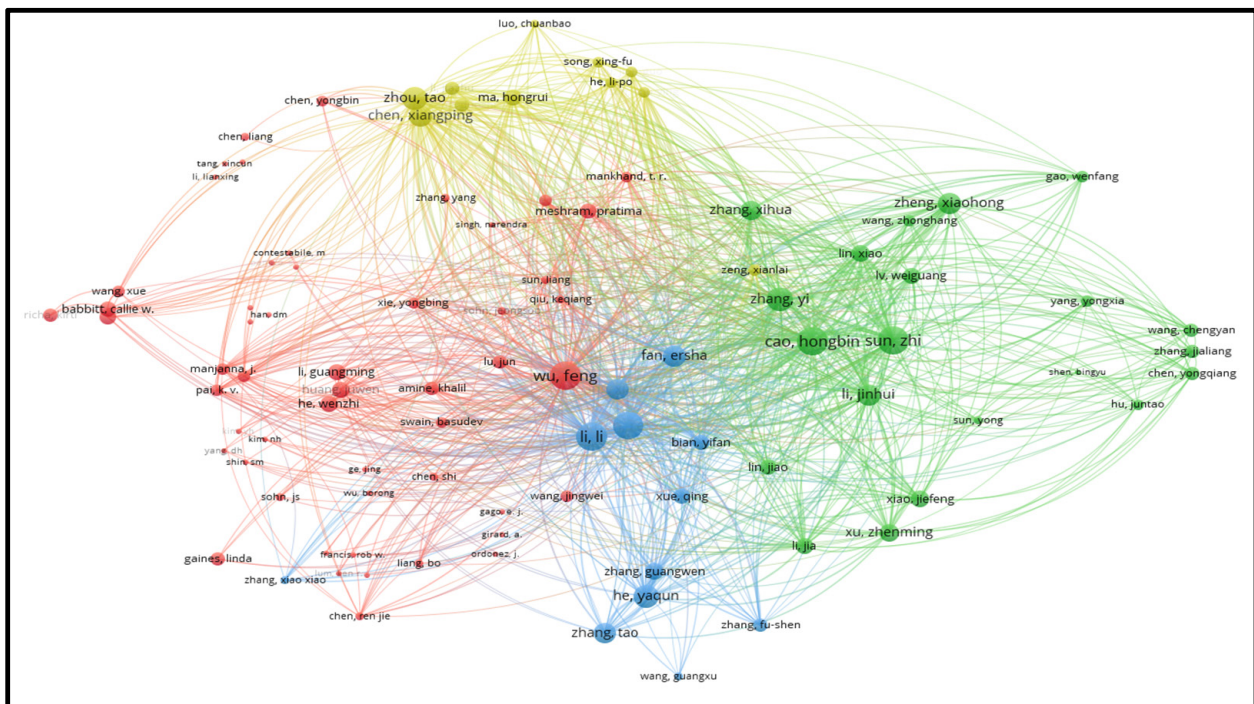


Figure 14. VOS Viewer Network Visualization of the 100 most cited authors.

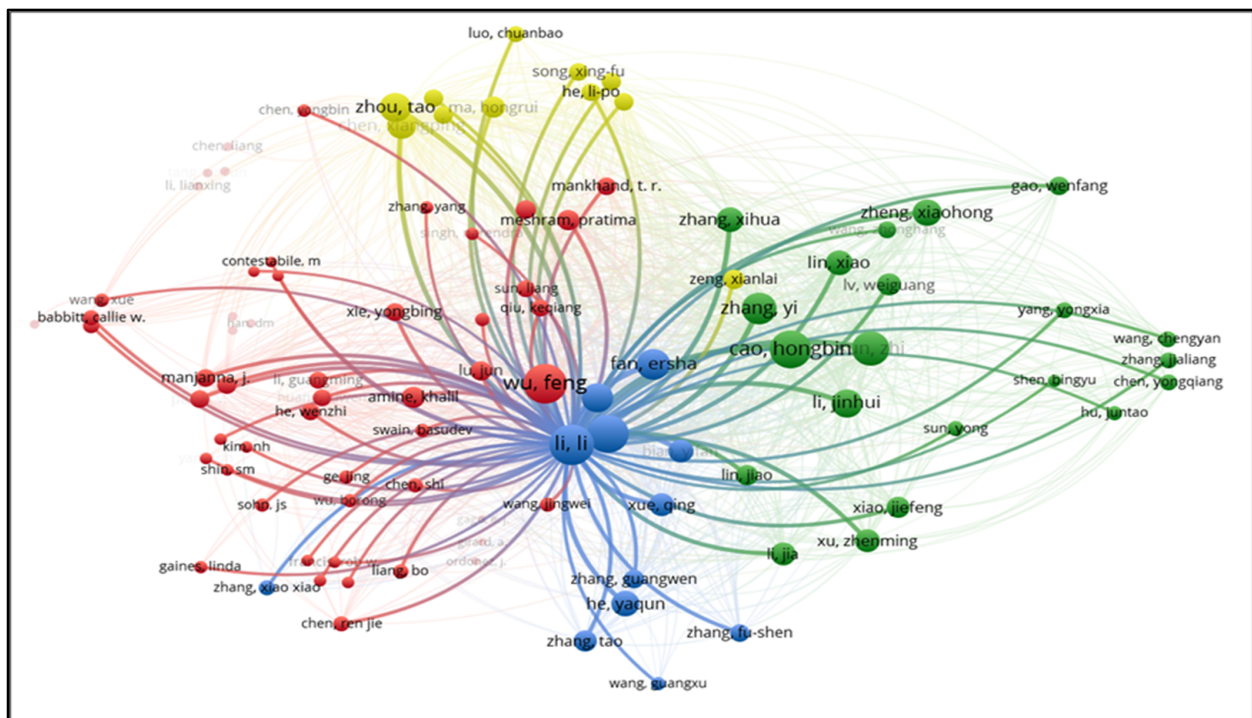


Figure 15. VOS Viewer Network Visualization Li, Li—Author with the greatest number of total citations.

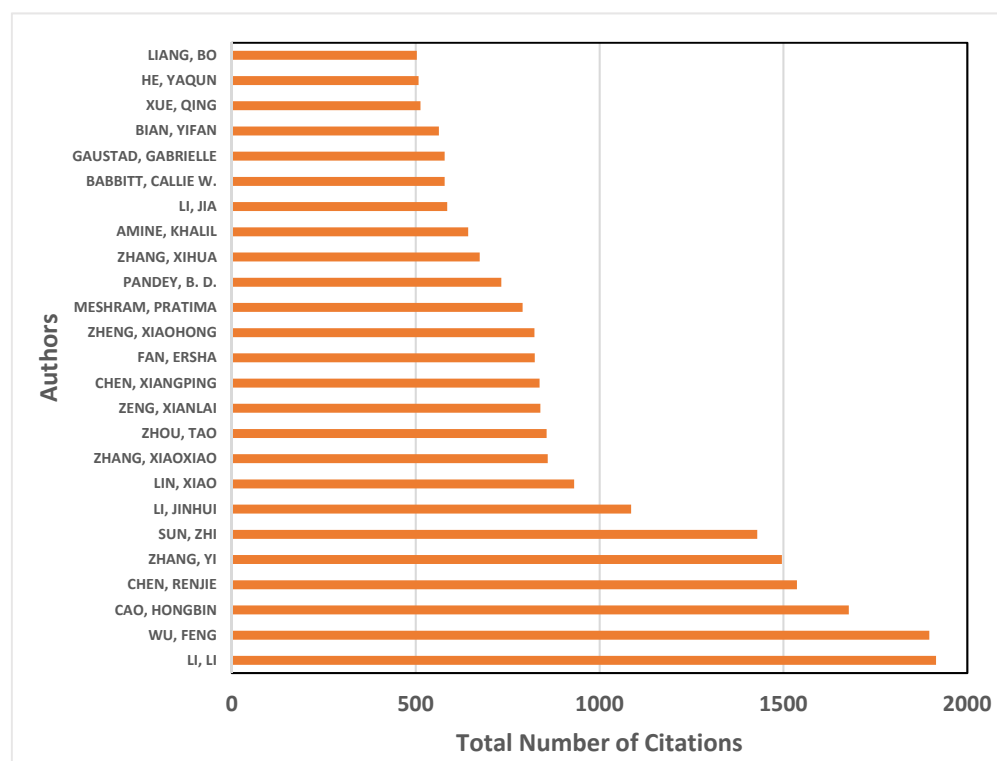


Figure 16. Authors with more than 500 total citations.

3.8. Average Number of Citations

Despite having the most citations overall and the largest number of publications, the authors on the top of the list do not tend to have the most relevant work when considering the number of average citations per article as shown in Figure 17.

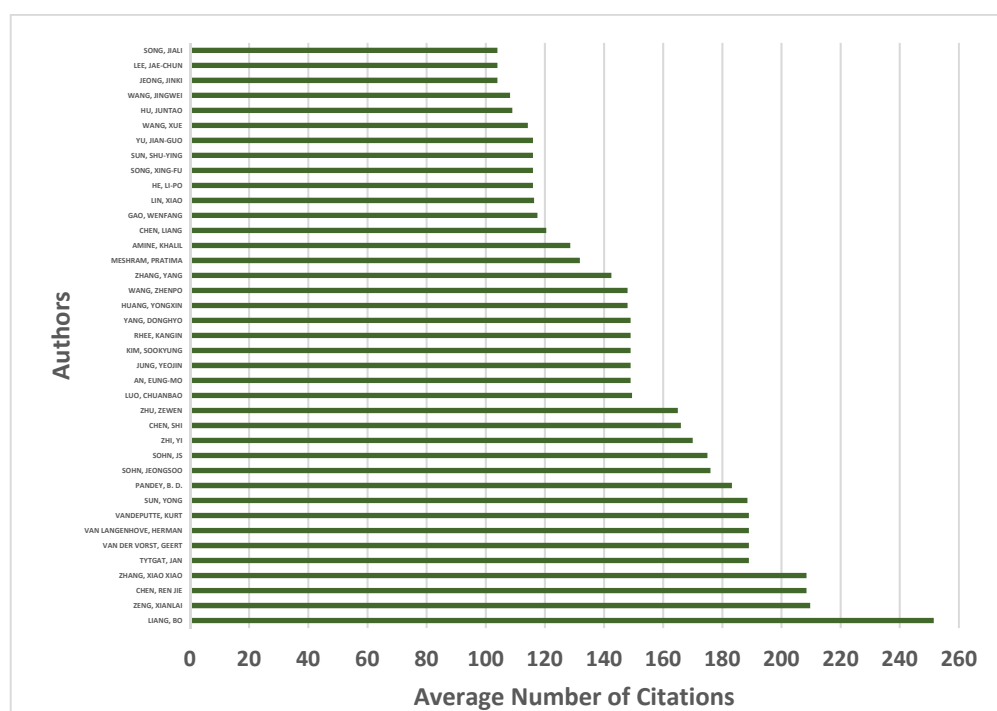


Figure 17. Authors with average citations greater than 100.

The metric, average number of citations, is extremely interesting because it shows how many times an author's work has been mentioned. In the case of Liang, Bo, number 1 on the list with only 2 published articles with a total of 503 citations for both articles averages 252 citations per article.

Both metrics are important to establish the most relevant and essential work in the LIB recycling field. The former shows authors whose numerous works throughout a period of time have contributed to the state-of-the-art in LIB research. The latter takes into account authors who, despite not having many published works, have articles which have skyrocketed and become a go-to article for the LIB recycling theme.

3.9. *Methodi Ordinatio* vs. *ProKnow-C*

The comparison between *Methodi Ordinatio* and *ProKnow-C* up to this point renders the same results when comparing the countries with the most publications, ranking of research groups, number of publications per year, number of articles published per journal, most common keywords, most published authors, most cited authors and authors with the best average citation. Nevertheless, when comparing the overall individual ranking of each article that composes the bibliographic portfolio based on the established metrics, there are divergences mainly in article position or ranking.

In the *Methodi Ordinatio*, out of the 591 articles which compose the BP, only 3 articles scored negatively meaning that these were either old articles or had very few citations. The other 588 articles are all considered relevant articles to the LIB recycling theme. Nevertheless, the idea of this BP is to establish a limited number of articles to which future researchers can consult, and to study and find out the state-of-the-art of LIB recycling without having to go through an immense amount of literature. With a means of reducing the BP, the Pareto Rule was applied, and the final *Methodi Ordinatio* BP came down to 175 articles.

To reduce the number of articles even further, an arbitrary metric was considered. The metric only lists articles whose *Ordinatio* score was at least 40% the value of the number 1 ranked article. The first nine articles listed in Table 4 have at least half the value of the number 1 article (50%) and articles ranked from the 10th to the 26th position have at least 40% of the value of the 1st article based on the *Ordinatio* ranking. These 26 articles compose the final BP based on the *Methodi Ordinatio*. Table 4 shows a ranking of the articles from the BP as well as the authors, and reference numbers.

Table 4. List of the highest-ranking articles based on the *Methodi Ordinatio*.

Rank	Title	Author	Ref.
1	Recovery and recycling of lithium: A review	Swain, B	[3]
2	A review of processes and technologies for the recycling of lithium-ion secondary batteries	Xu, JQ; Thomas, HR; Francis, RW; Lum, KR; Wang, JW; Liang, B	[6]
3	Recycling of Spent Lithium-Ion Battery: A Critical Review	Zeng, XL; Li, JH; Singh, N	[45]
4	Extraction of lithium from primary and secondary sources by pre-treatment, leaching and separation: A comprehensive review	Meshram, P; Pandey, BD; Mankhand, TR	[4]
5	A Critical Review and Analysis on the Recycling of Spent Lithium-Ion Batteries	Lv, WG; Wang, ZH; Cao, HB; Sun, Y; Zhang, Y; Sun, Z	[9]
6	Toward sustainable and systematic recycling of spent rechargeable batteries	Zhang, XX; Li, L; Fan, ES; Xue, Q; Bian, YF; Wu, F; Chen, RJ	[7]
7	Processes and technologies for the recycling and recovery of spent lithium-ion batteries	Ordenez, J; Gago, EJ; Girard, A	[5]
8	Recovery of cobalt and lithium from spent lithium ion batteries using organic citric acid as leachant	Li, L; Ge, J; Wu, F; Chen, RJ; Chen, S; Wu, BR	[46]

Table 4. Cont.

Rank	Title	Author	Ref.
9	Novel approach to recover cobalt and lithium from spent lithium-ion battery using oxalic acid	Zeng, XL; Li, JH; Shen, BY	[47]
10	Hydrometallurgical processing of spent lithium ion batteries (LIBs) in the presence of a reducing agent with emphasis on kinetics of leaching	Meshram, P; Pandey, BD; Mankhand, TR	[8]
11	Recovery of metals from spent lithium-ion batteries with organic acids as leaching reagents and environmental assessment	Li, L; Dunn, JB; Zhang, XX; Gaines, L; Chen, RJ; Wu, F; Amine, K	[48]
12	A promising approach for the recovery of high value-added metals from spent lithium-ion batteries	Hu, JT; Zhang, JL; Li, HX; Chen, YQ; Wang, CY	[49]
13	Sustainable Recycling Technology for Li-Ion Batteries and Beyond: Challenges and Future Prospects	Fan, ES; Li, L; Wang, ZP; Lin, J; Huang, YX; Yao, Y; Chen, RJ; Wu, F	[50]
14	Environmentally-friendly oxygen-free roasting/wet magnetic separation technology for in situ recycling cobalt, lithium carbonate and graphite from spent LiCoO ₂ /graphite lithium batteries	Li, J; Wang, GX; Xu, ZM	[51]
15	Process for the recovery of cobalt oxalate from spent lithium-ion batteries	Chen, L; Tang, XC; Zhang, Y; Li, LX; Zeng, ZW; Zhang, Y	[52]
16	Recovery of valuable metals from waste cathode materials of spent lithium-ion batteries using mild phosphoric acid	Chen, XP; Ma, HR; Luo, CB; Zhou, T	[53]
17	Development of a metal recovery process from Li-ion battery wastes	Shin, SM; Kim, NH; Sohn, JS; Yang, DH; Kim, YH	[2]
18	Lithium Carbonate Recovery from Cathode Scrap of Spent Lithium-Ion Battery: A Closed-Loop Process	Gao, WF; Zhang, XH; Zheng, XH; Lin, X; Cao, HB; Zhi, Y; Sun, Z	[54]
19	Organic oxalate as leachant and precipitant for the recovery of valuable metals from spent lithium-ion batteries	Sun, L; Qiu, KQ	[55]
20	Hydrometallurgical recovery of metal values from sulfuric acid leaching liquor of spent lithium-ion batteries	Chen, XP; Chen, YB; Zhou, T; Liu, DP; Hu, H; Fan, SY	[56]
21	Hydrometallurgical process for the recovery of high value metals from spent lithium nickel cobalt aluminum oxide based lithium-ion batteries	Joulie, M; Laucournet, R; Billy, E	[57]
22	A Mini-Review on Metal Recycling from Spent Lithium Ion Batteries	Zheng, XH; Zhu, ZW; Lin, X; Zhang, Y; He, Y; Cao, HB; Sun, Z	[34]
23	Ascorbic-acid-assisted recovery of cobalt and lithium from spent Li-ion batteries	Li, L; Lu, J; Ren, Y; Zhang, XX; Chen, RJ; Wu, F; Amine, K	[58]
24	Recycling of spent lithium-ion batteries in view of lithium recovery: A critical review	Liu, CW; Lin, J; Cao, HB; Zhang, Y; Sun, Z	[59]
25	Metals removal and recovery in bioelectrochemical systems: A review	Nancharaiah, YV; Mohan, SV; Lens, PNL	[60]
26	Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries	Sun, L; Qiu, KQ	[61]

The other method for evaluating the relevance of articles used was ProKnow-C. As was previously mentioned, the differences between *Methodi Ordinatio* and ProKnow-C come down to the ranking of the most relevant articles from the BP.

The ProKnow-C methodology initial database was composed of 591 articles just like *Methodi Ordinatio*. These articles were later filtered and selected through the Pareto Rule, where 147 articles (25%) met the threshold. The articles which were left out based on the

Pareto Rule (444 total) went through other analyses to see if they could be incorporated into the final BP. The first step was to analyze the year of publication. If these articles were considered to be new or recent (publication from 2017 on), they would be extricated from exclusion. By doing so, 394 articles (67%) were introduced into the BP. The final step was to rescue articles which were either left out by the Pareto Rule or were not considered recent. The relevance of these articles is due to the authors. Therefore, authors whose articles had not made the first and second cut would now be reintroduced to the BP. In this step, only 19 articles (3%) were reinserted into the BP, making a total of 560 selected articles for the final ProKnow-C BP. These articles were ranked based on their final score, which considered the number of citations, JCR, age, and selection step (Pareto, age, or author recognition phase).

When comparing *Methodi Ordinatio* and ProKnow-C, there is a clear difference in the final BP. Even though both start with 591 articles, after applying the Pareto Rule, *Methodi Ordinatio* BP is left with 175 articles, while ProKnow-C is left with 147. These articles are the ones considered relevant to the research theme based on the Pareto Rule. The other articles reinserted by ProKnow-C based on their age and author recognition becomes too broad and the number of articles in the database is essentially what was started with (560 articles). Despite being a necessary step in the ProKnow-C methodology, these steps did not influence the final BP, since no articles from these phases made it into the top 50.

To reduce the BP based on the ProKnow-C methodology from 147 articles to about the same number as the *Methodi Ordinatio*, an arbitrary metric was also applied. In doing so, a list of 23 best-ranked articles was established. The same metric applied to *Methodi Ordinatio* was also applied to ProKnow-C. Only articles whose value was 40% the value of the 1st article's score would be considered. A list of the best-ranked articles based on the ProKnow-C Methodology is shown in Table 5.

Table 5. List of the highest-ranking articles based on the ProKnow-C Methodology.

Rank	Title	Author	Ref.
1	Recovery and recycling of lithium: A review	Swain, B	[3]
2	A review of processes and technologies for the recycling of lithium-ion secondary batteries	Xu, JQ; Thomas, HR; Francis, RW; Lum, KR; Wang, JW; Liang, B	[6]
3	Recycling of Spent Lithium-Ion Battery: A Critical Review	Zeng, XL; Li, JH; Singh, N	[45]
4	Extraction of lithium from primary and secondary sources by pre-treatment, leaching and separation: A comprehensive review	Meshram, P; Pandey, BD; Mankhand, TR	[4]
5	Recovery of cobalt and lithium from spent lithium ion batteries using organic citric acid as leachant	Li, L; Ge, J; Wu, F; Chen, RJ; Chen, S; Wu, BR	[46]
6	Development of a metal recovery process from Li-ion battery waste	Shin, SM; Kim, NH; Sohn, JS; Yang, DH; Kim, YH	[2]
7	A laboratory-scale lithium-ion battery recycling process	Contestabile, M; Panero, S; Scrosati, B	[1]
8	A Critical Review and Analysis on the Recycling of Spent Lithium-Ion Batteries	Lv, WG; Wang, ZH; Cao, HB; Sun, Y; Zhang, Y; Sun, Z	[9]
9	Recovery of metal values from spent lithium-ion batteries with chemical deposition and solvent extraction	Nan, JM; Han, DM; Zuo, XX	[62]
10	Processes and technologies for the recycling and recovery of spent lithium-ion batteries	Ordonez, J; Gago, EJ; Girard, A	[5]
11	Process for the recovery of cobalt oxalate from spent lithium-ion batteries	Chen, L; Tang, XC; Zhang, Y; Li, LX; Zeng, ZW; Zhang, Y	[52]
12	Novel approach to recover cobalt and lithium from spent lithium-ion battery using oxalic acid	Zeng, XL; Li, JH; Shen, BY	[47]

Table 5. *Cont.*

Rank	Title	Author	Ref.
13	Recovery of metals from spent lithium-ion batteries with organic acids as leaching reagents and environmental assessment	Li, L; Dunn, JB; Zhang, XX; Gaines, L; Chen, RJ; Wu, F; Amine, K	[48]
14	Toward sustainable and systematic recycling of spent rechargeable batteries	Zhang, XX; Li, L; Fan, ES; Xue, Q; Bian, YF; Wu, F; Chen, RJ	[7]
15	Hydrometallurgical processing of spent lithium ion batteries (LIBs) in the presence of a reducing agent with emphasis on kinetics of leaching	Meshram, P; Pandey, BD; Mankhand, TR	[8]
16	Organic oxalate as leachant and precipitant for the recovery of valuable metals from spent lithium-ion batteries	Sun, L; Qiu, KQ	[55]
17	Recovery of cobalt sulfate from spent lithium ion batteries by reductive leaching and solvent extraction with Cyanex 272	Kang, J; Senanayake, G; Sohn, J; Shin, SM	[63]
18	Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries	Sun, L; Qiu, KQ	[61]
19	Ascorbic-acid-assisted recovery of cobalt and lithium from spent Li-ion batteries	Li, L; Lu, J; Ren, Y; Zhang, XX; Chen, RJ; Wu, F; Amine, K	[58]
20	Hydrometallurgical process for the recovery of high value metals from spent lithium nickel cobalt aluminum oxide based lithium-ion batteries	Joulie, M; Laucournet, R; Billy, E	[57]
21	Environmentally friendly oxygen-free roasting/wet magnetic separation technology for in situ recycling cobalt, lithium carbonate and graphite from spent LiCoO_2 /graphite lithium batteries	Li, J; Wang, GX; Xu, ZM	[51]
22	Recycling rechargeable lithium ion batteries: Critical analysis of natural resource savings	Dewulf, J; Van der Vorst, G; Denturck, K; Van Langenhove, H; Ghyoot, W; Tytgat, J; Vandeputte, K	[64]
23	A promising approach for the recovery of high value-added metals from spent lithium-ion batteries	Hu, JT; Zhang, JL; Li, HX; Chen, YQ; Wang, CY	[49]

Observing Tables 4–6, a difference in article ranking can be seen. Only the first four articles remain in the same position, which establishes these articles as a must-read for new researchers. Despite using two different methodologies, these top 4 articles still manage to hold their place based on the number of citations, relevance, and year of publication. The other articles vary in position based on the methodology, chosen as shown in Table 6.

Table 6. Rank comparisons between ProKnow-C and *Methodi Ordinatio*.

Title	ProKnow-C Ranking	Methodi Ordinatio Ranking
Recovery and recycling of lithium: A review	1	1
A review of processes and technologies for the recycling of lithium-ion secondary batteries	2	2
Recycling of Spent Lithium-Ion Battery: A Critical Review	3	3
Extraction of lithium from primary and secondary sources by pre-treatment, leaching and separation: A comprehensive review	4	4
Recovery of cobalt and lithium from spent lithium ion batteries using organic citric acid as leachant	5	8

Table 6. Cont.

Title	ProKnow-C Ranking	Methodi Ordinatio Ranking
Development of a metal recovery process from Li-ion battery wastes	6	17
A laboratory-scale lithium-ion battery recycling process *	7	52
A Critical Review and Analysis on the Recycling of Spent Lithium-Ion Batteries	8	5
Recovery of metal values from spent lithium-ion batteries with chemical deposition and solvent extraction *	9	36
Processes and technologies for the recycling and recovery of spent lithium-ion batteries	10	7
Process for the recovery of cobalt oxalate from spent lithium-ion batteries	11	15
Novel approach to recover cobalt and lithium from spent lithium-ion battery using oxalic acid	12	9
Recovery of metals from spent lithium-ion batteries with organic acids as leaching reagents and environmental assessment	13	11
Toward sustainable and systematic recycling of spent rechargeable batteries	14	6
Hydrometallurgical processing of spent lithium ion batteries (LIBs) in the presence of a reducing agent with emphasis on kinetics of leaching	15	10
Organic oxalate as leachant and precipitant for the recovery of valuable metals from spent lithium-ion batteries	16	19
Recovery of cobalt sulfate from spent lithium ion batteries by reductive leaching and solvent extraction with Cyanex 272 *	17	35
Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries *	18	26
Ascorbic-acid-assisted recovery of cobalt and lithium from spent Li-ion batteries	19	23
Hydrometallurgical process for the recovery of high value metals from spent lithium nickel cobalt aluminum oxide based lithium-ion batteries	20	21
Environmentally-friendly oxygen-free roasting/wet magnetic separation technology for in situ recycling cobalt, lithium carbonate and graphite from spent LiCoO ₂ /graphite lithium batteries	21	14
Recycling rechargeable lithium ion batteries: Critical analysis of natural resource savings *	22	46
A promising approach for the recovery of high value-added metals from spent lithium-ion batteries	23	12

Out of the 23 articles listed by the ProKnow-C methodology as the most relevant, 5 articles do not make up the 23 most relevant based on the *Methodi Ordinatio*. These articles are highlighted with an asterisk (*) in Table 6.

Despite not having a huge impact on previous analyzes, the *Methodi Ordinatio* vs. ProKnow-C investigation differentiates on article ranking seeing as they both consider different metrics and values when establishing a ranking. Another interesting observation is that articles that overlap are the ones that make up the most relevant state-of-the-art published work.

4. Conclusions

This review article showed that by using two distinct methodologies, ProKnow-C and *Methodi Ordinatio*, it was possible to establish a systematic analysis of 2515 articles found in two search engines: SCOPUS and WoS. After initial filtering and evaluation, 591 articles were left in each database (ProKnow-C database and *Methodi Ordinatio* database). There was no differences found in these articles, which means that the 591 articles in *Methodi Ordinatio* were the same as the ones found in ProKnow-C.

The analysis of these 591 articles made it possible to compare the results between both methods used, ProKnow-C and *Methodi Ordinatio*. The overall view indicated no difference, meaning that up to a certain point the results regarding the BP rendered the same information for both ProKnow-C and *Methodi Ordinatio*. The similar results indicated that the People's Republic of China (40.6%) and the USA (12.6%) have the greatest number of publications in the LIB recycling field. Other results that showed the same outcome for both methods include: research group affiliation, publications per year, publications per journal, keywords, number of published articles, author citation and average number of citations per author.

The significant difference in both methodologies could be seen in the ranking of the articles, which were used to build a concise and relevant BP. Two lists of the best 23 ranked articles were formed, one for each methodology. When comparing the best ranked articles from ProKnow-C BP with *Methodi Ordinatio* BP, the only similarity is that the top four ranked articles were the same for both databases. The other 14 articles were positioned in different ranks while still making the cut of the best 23 ranked. There were 5 articles from each BP which did not make the cut, designating that they were ranked after the 23rd position.

This result indicates that despite their similarity in the number of phases and parameters, ProKnow-C and *Methodi Ordinatio* can bring about different results. Hence, the importance of comparing and contrasting the two methodologies, which allows for a more precise final BP in the sense that articles that are ranked in the same position, or close positions in both ProKnow-C and *Methodi Ordinatio* become undoubtedly reference articles for the desired theme. In this review article, a list of the 18 fundamental articles for understanding the state-of-the-art in LIB recycling was produced.

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