# Technological change in lithium iron phosphate battery: the key-route main path analysis

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**Abstract** Technological change evolves along a cyclical divergent-convergent pattern in knowledge diffusion paths. Technological divergence occurs as a breakthrough innovation, or discontinuity, inaugurating an era of ferment in which several competing technologies emerge and gradually advance. Technological convergence occurs as a series of evolutionary, variant changes that are gradually combined or fused together to open the industry to successive dominant designs or guideposts. To visualize such a pattern of technological evolution, we choose to study lithium iron phosphate (LFP) battery technology through an extension of the citation-based main path analysis, namely the key-route main path analysis. The key-route method discloses the main paths that travel through a specified number of key citations. The resulting multiple paths reveal the structure of the knowledge diffusion paths. The citation network is constructed from 1,531 academic articles on LFP battery technology has completed two full technological cycles and is in the middle of the third cycle.

**Keywords** Technological evolution · Technological cycle · Main path analysis · Lithium iron phosphate battery

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## Introduction

Sir Isaac Newton once remarked: "If I have seen further it is by standing on the shoulders of giants." This remark highlights a fact that scientific researchers and innovators develop new ideas based on the knowledge of others. Knowledge is improved, extended, and combined continuously in the course of its development. The chains in which knowledge disseminates and recombines are commonly called knowledge flow, or knowledge diffusion paths.

The phenomena of knowledge diffusion have been studied extensively in the innovation, science, and technology literature. However, most of this work has focused on knowledge spillovers within or across organizational boundaries (Breschi and Lissoni 2001; Singh 2005), rather than on the role of knowledge diffusion in technological innovation processes. Besides, no studies have elaborated on the topographical pattern of knowledge diffusion paths.

Several recent studies on innovation have begun to use knowledge diffusion paths to trace technological trajectories (Verspagen 2007; Mina et al. 2007; David et al. 2011; Martinelli 2012; Bhupatiraju et al. 2012). Through a new approach, these studies are able to map or visualize the most significant knowledge diffusion paths of a target technology and match the flow of knowledge on visualized paths with the observed technological changes. In other words, they found that the main flow of knowledge matches well with the technological trajectories. More interestingly, through observing disruptions of knowledge diffusion paths, Martinelli (2012) indicates that the approach has potential to detect patterns of technological changes.

Following this line of thought, we illustrate that the topographical pattern in knowledge diffusion paths can indeed be used to distinguish paradigmatic and trajectory changes. More specifically, we suggest that knowledge diffuses topologically in a repetitive divergent-convergent pattern in the development of a technology and that divergence occurs immediately after a technological breakthrough, while convergence begins after a standard design is settled and ends at another breakthrough, which begins the next cycle.

A repetitive divergent-convergent pattern denotes that knowledge diffuses in cycles, wherein each phase is made up of a knowledge dispersion period followed by a knowledge concentration period. This notion of divergent-convergent patterns in knowledge diffusion is closely related to the way a technology evolves (Anderson and Tushman 1990). To be more specific, the pattern is a consequence of shifts in paradigms and trajectories. As such, we use the topological pattern of knowledge diffusion paths to detect technological changes; in other words, to define technological trajectories in terms of knowledge flows (Martinelli 2012).

The problem of recognizing knowledge diffusion pattern is in a way similar to the problem of human activity pattern analysis (Hui et al. 2009; D'Urso and Massari 2013). Like knowledge diffusion, some human activities such as grocery shopping, web browsing, etc. form ordered sequences. Human activity pattern analysis is rooted in a well-established discipline—sequence comparison. The techniques developed in the sequence comparison discipline have long been applied to areas such as speech processing, homology of DNA, handwriting analysis, and text collation, etc. (Kruskal 1983a, b; Pearson and Lipman 1988). It is tempting to use the techniques developed in sequence comparison discipline to address our problem. There is, however, a major difference between our problem and the sequence comparison one. In a typical sequence comparison or human activity pattern analysis problem, the movement of a sequence can return to the same identity (such as a shop, a web page, or a nucleotide) repetitively, yet knowledge diffuses only forward and never flows back to where it had passed through. The techniques developed cannot be applied directly to our problem. We thus resort to the traditional bibliometric method—citation analysis.

Citation analysis has long been used as a tool in journal evaluation (Garfield 1972), where citation frequency provides a measure of the impact of a journal. In addition to counting frequency, citation analysis also examines the connections among publications. This is usually done through establishing a citation network. A citation network is an unweighted directional network in which the nodes represent publications and the links among nodes represent citation relationships. We draw on the works of Hummon and Doreian (1989) and Verspagen (2007) on the connectivity analysis of citation networks commonly located under the name of "the main path analysis."

Hummon and Doreian (1989) suggested a method to determine the significant citation links in a citation network and then connect some of these links to form a single sequence of significant citation links, which they call the "main path." They demonstrated the method using a citation network made up of a small set of DNA papers. The papers on the main path were also identified to be the most important papers by other researchers who applied a completely different methodology. The single main path uncovered through the method provides a crucial part of the governing structure. However, describing the development of a scientific field or technology in just a single path runs the risk of missing some elements of the governing structure, not to mention that the approach itself is not guaranteed to include the most significant citation link in the single main path.

Based on Hummon and Doreian (1989); Verspagen (2007) endeavored to map technological trajectories of fuel cell technology through patent citation network. Rather than examining only one single main path, they adopted a slightly different approach which allows the presence of multiple sequences of citation links. These multiple main paths are very close in revealing the governing structure, but fall short of not knowing whether the top significant citation links are included.

Bhupatiraju et al. (2012), in an investigation of knowledge flows in the core literature of innovation, entrepreneurship, and technology studies, applied the same main path approach as Verspagen (2007). They observed many situations of knowledge flow convergence and divergence from the topology of the main paths. Martinelli (2012) also applied a similar approach to Verspagen (2007) to examine the technological changes in the telecommunications switching industry. In addition to the main path method (quantitative), the study used changes in engineering heuristics (qualitative) to detect paradigmatic shift. Such a combination of quantitative and qualitative approaches successfully observed paradigmatic shifts that match what have been identified previously.

Unlike the works mentioned, we proposed using key-route main paths (Liu and Lu 2012) to examine technological changes from the topology of knowledge diffusion paths. Key-route main paths are multiple main paths constructed based on key-routes, which are the top significant links in a citation network. Key-route main paths are different from the main path suggested by Hummon and Doreian (1989); the former includes multiple sequences of citations from a set of starting points to another set of ending points, whereas the latter only refers to a single sequence of citation. Furthermore, key-route main paths are different from the multiple main paths obtained by Verspagen (2007), in that the key-route paths guarantee that the top significant citation links in the multiple sequences reveals the governing structure of the knowledge diffusion paths and allows the divergent-convergent pattern to surface, if it exists.

Our research target is lithium iron phosphate (LiFePO4, or LFP) battery technology, from which we construct a set of academic papers to examine the citation paths. We chose publications as a proxy for measuring technological change mainly for two reasons. First, we chose to analyze academic papers because—when compared with other possible proxies such as R&D spending and patents—they are better matched with our research focus on using

knowledge diffusion paths to trace technological trajectories. Second, because technological cycle time is measured by the time span between the predecessor and the successor, we need a proxy that is likely to develop into a path-dependent evolutionary process, especially over a long time span. Academic publications meet this requirement. In the case of LFP battery development, after the main design concept was proposed in an academic paper (Padhi et al. 1997) the papers that followed closely recorded the historical movement of the changes in design concepts. The patents on the LFP battery technology, on the other hand, do not contain such rich information, especially since patent citation is usually sparser than paper citation. In addition, owing to a faster review cycle, academic papers reflect the state-of-the-art faster than patents. Our choice is generally consistent with Thomke and Kuemmerle (2002), who have likewise used scientific publications as a proxy for measuring technological change in combinatorial chemistry and high-throughput screening.

The remaining portion of this article is organized as follows: the next section introduces the research background, including research in technological divergence-convergence, the key-route main path analysis, and the LFP battery technology; Sect. 3 details the analytical procedures of the key-route path method that we adopted; Sect. 4 describes the data sources and presents the knowledge diffusion paths of LFP battery technology; and the final section discusses the limitations and contributions of the study, as well as implications of the results.

## Background

The literature of science and technology evolution studies has long recognized the pathdependent and cyclical nature of technological change or innovation in industry. Accounts differ, but generally, a pattern of repeated chaotic and structured activities is seen to define particular areas of technology that evolve over time (Anderson and Tushman 1990; Dosi 1982; Hung 2004; Levinthal 1998; Nelson and Winter 1982).

Three central themes on evolution are noted here. First, a technological breakthrough will trigger a chaotic period of design competition. The breakthrough concept is then branched off into multiple competing designs. Second, one design eventually wins over other designs and is accepted as the standard—a "dominant design" (Abernathy and Utterback 1975) or "technological guidepost" (Sahal 1985). Third, the standard guides the technological development for a period of time when there is only incremental progress, and the competing designs gradually fuse together to a point when the period ends with another breakthrough. A technology evolving as successive cycles is usually sustainable, either because it is supported by external technologies (Kodama 1992) and institutions (Lundvall 1992), or because it produces internal externalities (Arthur 1989).

Divergent-convergent patterns of knowledge diffusion paths

These process-oriented themes indicate the possibility of determining knowledge flow in a phase transition characterized by repetitive divergence-convergent links. Because technological change is best detected at knowledge levels (Martinelli 2012; Verspagen 2007), a path dependent, cyclical model of technological change, such as the previously mentioned model, is consistent with a divergent-convergent pattern of knowledge diffusion paths.

More specifically, the chaotic or discontinuous period is full of new knowledge that branches off from the initial breakthrough concept. The new knowledge may, in turn, spawn a new set of knowledge as technology evolves. The amount of new knowledge, or the number of the new studies, grows rapidly, but this new knowledge can all be traced diffuses divergently to a point when the design standard is surfaced. At this point, the momentum of growth is reduced and the innovation process is phased into incremental progress periods, where fewer ideas are proposed. The existing knowledge is slowly combined together or discarded. The period ends when another major breakthrough concept is proposed. The new breakthrough concept is built upon several extensions of the previous knowledge. One thus expects that the previous knowledge converges with this new breakthrough concept. As technology evolves, the repetitive divergence-convergence pattern in knowledge diffusion paths prevails over other patterns.

Figure 1 depicts such a divergent-convergent pattern of knowledge diffusion paths. This figure presents two full stages of divergent-convergent cycles, each beginning with a breakthrough and ending in another breakthrough, plus a divergent half cycle. The divergent phase corresponds to the period of design competition, and the convergent phase is characterized by the elaboration of a dominant design. In between is the peak of the chaotic period, which is also the time when the dominant design begins to surface.

The concept of a divergent-convergent pattern in knowledge diffusion paths has not previously been discussed or examined in the literature. The closest study is Bhupatiraju et al. (2012), which repeatedly used the words "converge" and "diverge" to describe the mergence and divergence of knowledge flows. They observe that multiple knowledge paths "converge on each other" (p. 1211) and therefore, state that "the convergence-divergence dynamic of main paths will be a main topic in the discussion of our results" (p. 1211). Despite this acknowledgement, Bhupatiraju et al. (2012) did not further explore in their study how this phenomenon of the convergence-divergence dynamic of main paths actually comes into function during the paths of evolution.

#### The key-route main path analysis

To address the gap in mapping a cyclical model of technological change at the knowledge level, we apply the key-route main path analysis to visualize the key knowledge diffusion paths of a particular technology. As mentioned in the introductory section, our goal is to uncover the governing structure of knowledge diffusion paths. Only with multiple paths can we visualize the interwoven paths, and only with the inclusion of the top significant citation links can we regard the interwoven paths' governing structure.

The key-route main path analysis extends the methods of Hummon and Doreian (1989) and Verspagen (2007) by adding an algorithm to search for multiples paths and guaranteeing the inclusion of the top significant links in these multiple paths (Liu and Lu 2012). Improved over the single path suggested by Hummon and Doreian (1989), the key-route main path analysis provides multiple paths. Unlike Verspagen (2007), which may not catch all the most significant links in their paths, the key-route main path analysis includes all the top most significant links in the multiple paths.

In summary, the main path analysis with the key-route approach is an effective tool to detect technological changes. Based on the assumption that the technological changes are embedded in the governing structure of the knowledge diffusion paths, once the governing structure is made to surface through the key-route approach, the stories of technological changes speak for themselves.

We define a key-route as a significant citation link in the citation network. For a given number of key-routes, the analysis finds a collection of paths, each travelling through one of the specified key-routes. These key-route paths interweave with each other so that when they are merged together, the collection of paths reduces to one knowledge diffusion map



Fig. 1 Technological cycle as a divergent-convergent linkage

that includes all the specified links. When the specified links are the top most significant citations in the citation network, this knowledge diffusion map not only contains much richer information than the traditional single knowledge path, it also illustrates the governing structure of the knowledge flow. It is expected that this governing knowledge flow structure will reveal how technology changes in its development. In addition, depending on the number of key-routes specified, one is able to examine different level of details of the structure. The higher the number of key-routes, the more structural detail can be visualized.

## LFP battery technology

Our study target is LFP battery technology, a central emerging technology for green vehicles. In view of energy efficiency, green vehicles produce less harmful impacts to the environment than conventional vehicles running on gasoline or diesel. The demand for green vehicles has grown considerably under increasing concerns of carbon dioxide emissions and rising gasoline prices in recent years. A battery electric vehicle (BEV) is an important type of green vehicle that uses chemical energy stored in rechargeable battery packs. Among the battery technologies available for a BEV, the LFP battery has gained much attention due to its high energy density, low raw materials cost, environmental friendliness, and safety (Chung et al. 2002).

We studied LFP battery technology for two particular reasons. First, the technology exhibits the characteristic of a new scientific and technology field—a very clear starting point. The development of the technology began from a breakthrough in 1997, when Professor Goodenough's research group at the University of Texas at Austin proposed to use phospho-olivines as cathode materials for rechargeable lithium batteries (Padhi et al. 1997). The breakthrough inspired a group of researchers and spawned a sequence of investigations that worked within the framework of the original concept.

Second, the technology is under heavy competition and changes dynamically, considering the fact that researchers in this field of technology have published more than 1,500 papers in only 15 years. According to the concept of a paradigm shift, there is a better chance for us to see a divergent-convergent pattern in a technology with heavy changes during its development. Therefore, we apply the key-route main path analysis on LFP to verify the existence of divergent-convergent patterns.

#### Analytical procedures

The main path analysis traces knowledge diffusion pathways by examining citation information embedded in scientific publications. When the publication of an author cites a

work of his or her peers, the author has presumably considered that the concepts in the peers' work exert some influence in the paper he or she is now publishing. Certain knowledge is diffused from the cited work to the author's publication through a virtual knowledge conduit between the two publications. Knowledge can disseminate downward through such conduits for several generations. A chain of knowledge conduits forms a knowledge diffusion path. For each publication, there can be numerous knowledge diffusion paths emanating from it.

The first step in tracing knowledge diffusion paths in a scientific field is to construct a citation network based on citation relationships in the publications within the field. Because knowledge flows from a cited publication to the citing publications, we select to direct the link between two nodes from the cited paper to the citing paper.

In a citation network, a "source" is a node that is cited, but cites no other nodes; a "sink" is a node that cites other nodes, but is not cited. In other words, sources are the origins of knowledge, while sinks are the endpoints of knowledge dissemination. Nodes other than sources and sinks are referred to as intermediate nodes. Our interest here is to trace knowledge diffusion paths that begin at sources and end at sinks. In a normal citation network, there are usually a significant number of such paths; they increase exponentially with the total number of nodes in a citation network.

It would be difficult to make sense of such a large number of knowledge diffusion paths without some differentiation techniques. Hummon and Doreian (1989) first proposed methods to assign a weight to each citation link. Batagelj (2003) suggested a similar algorithm called search path count (SPC), which is the method we are adopting here. A citation link's SPC is the number of times the link is traversed if one exhausts the search from all the sources to all the sinks in a citation network. A link with a large SPC indicates that it sits on a large number of knowledge diffusion paths and, therefore, has to have a certain importance in the knowledge dissemination process.

We use the example citation network—shown in Fig. 2a—to demonstrate how SPCs for each link are calculated. The network includes 10 nodes; among them are two sources, A and B, and four sinks, F, H, I, and J. If one exhausts searching all paths from all the sources to all the sinks, then link C-E is passed through by four paths: A-C-E-H, A-C-E-J, B-C-E-H, and B-C-E-J. Its SPC value is therefore 4. Link D-G's SPC value is 2, because it is traversed by two paths: B-D-G-I and B-D-G-J. In the example network, links C-E and B-D have the largest SPC value.

After the SPC value for each citation link is calculated, the main path of a citation network—as suggested in Verspagen (2007)—is identified through the following procedures. First, determine the link with the largest SPC from all possible links emanating from the sources. Assign the beginning node of this link as the start point of the main path. Take the ending node of the link as the starting point for the next step. If there are ties, take all the tied links into consideration.

Second, determine the link with the largest SPC emanating from the current start point(s). Take the ending node(s) of the link(s) as the start point(s) for the next step. If the ending node is a sink, stop. If there are ties at each start point, take all the tied links into consideration. Finally, continue with the second step until all the paths hit a sink.

Following these steps, one finds the main path(s) of the example network to be B-D-G-I and B-D-G-J. The main path(s) identified in this fashion are what we call the "local" main path, as it always selects the current route with the largest SPC value, but the overall (accumulated) SPC value of this local main path is not the largest among all paths. For example, the overall SPC value of the paths A-C-E-H, A-C-E-J, B-C-E-H, and B-C-E-J is 9,



**Fig. 2** An example citation network (**a**) and its corresponding main paths (**b**, **c**, **d**). The numbers attached with the links are their SPC values. Figure **b** and **c** present the local and global main paths, respectively. Figure **d** represents the key-route main paths based on four links with the largest SPC value. The main paths are marked with bold lines

which is bigger than that of the local main paths at 7. The significance of these four paths should not be ignored.

These paths were first used in Verspagen (2007) and were referred to as "top paths" in Martinelli (2012) and David et al. (2011). We refer to these paths as "global" main paths, in contrast to "local" main paths. The global main path emphasizes the overall importance, while the local main path highlights the progressing significance. Figures 2b and c illustrate the local and global main paths of the example network, respectively. The problem of finding the path with the overall largest SPC value is similar to the shortest path problem in graph theory (Floyd 1962). For a citation network, which is always acyclic, several algorithms to solve the problem are readily available.

While the global and local main paths present the paths of significance, one finds that the link(s) with the largest SPC value may not always be included. For example, link B-D is missing in the global main path and C-E is missing in the local main path. The concept of key-route main paths is designed to overcome this issue.

The key-route main path is constructed as follows: select the link(s) with the largest SPC value as the key-route(s); search forward from the end node of the key-route(s) until a sink is hit; and search backward from the start node of the key-route(s) until a source is hit. The forward and backward searching can be accomplished through either local or global methods.

The procedure guarantees that the desired links (with the largest SPC value) are included in the main paths. A nice feature of the scheme is that one is able to "zoom" in or out to examine different levels of detail by selecting the number of key-routes. The more key-routes are selected, the more detail of the paths is observed. The results of key-route main paths usually contain both the local and global main paths. They are very useful in demonstrating the structural pattern of knowledge diffusion paths. In this study, we use the key-route main paths to illustrate the divergent-convergent pattern.

Figure 2d presents the key-route main paths constructed based on the four links with the largest SPC value: B-D, C-E, A-C, and B-C. Searching forward and backward from these links, we obtained the key-route main paths: A-C-E-H, A-C-E-J, B-C-E-H, B-C-E-J, B-D-G-I, and B-D-G-J.

## Data and results

#### Data sources

We collected LFP battery technology papers from the ISI Web of Science (WOS) database on April 4, 2012. The starting year was set to 1997, when Padhi et al. (1997) published their groundbreaking work. In searching the papers, we first referred to several review articles in the field (Fergus 2010; Yuan et al. 2011; Zhang 2011; Wang et al. 2011) to extract a set of commonly used keywords. We then applied the Boolean search technique, suggested by Porter et al. (2008), to improve the accuracy and precision of the search.

The search for LFP battery technology papers is separated into five steps. First, we find all papers with topic (TS) related to LFP and the family, and denote the resulting dataset as #1. In WOS, TS indicates searching in the title, abstract, author keywords, or Keywords Plus<sup>®</sup>. The search terms in this step include "lithium iron phosphate\*", LiFePO4, LiM-PO4, LiMnPO4 LiFe1-x Mn (x) PO4, etc. Second, we refine #1 to make sure that the dataset includes only papers associated with the structure, cathode, battery, and electrochemical properties, and denote the results as #2. The additional search terms include "olivine\*", "structure\*", "crystal\*", "batter\*", "cell\*", etc. The third step is to put back papers that may be overkilled in the previous step. This is done by applying the same set of keywords as in Step 1 to do the search, but only on title (TI). After combining the results of this step with #2 we obtain the dataset #3. Fourth, we further identify a set of papers that contains keywords in the paper title which are similar to, but not associated with, LFP. This dataset is denoted as #4. The search terms in this step include "Li\*O2", "LiMn0.33-Ni0.33Co0.33O2", "LiMn2O4", "LiVOPO4", "Li2Ni2(MoO4)(3)", "lithium iron orthosilicate", etc. In the end, we obtain the final dataset by removing those papers in dataset #4 from #3. This preliminary dataset consists of 1,531 academic papers.

Citation information for each paper in the preliminary dataset was then retrieved from the WOS database one by one. Before proceeding further, this information was used to fine-tune the dataset. Those papers that received no citations and did not cite other papers in the dataset were excluded, so as to reduce the total number of papers to 1,480. This is the dataset we used to construct the citation network. In the citation network, there were a total of 17,015 links that connected 39 sources, 492 sinks, and 949 intermediate nodes. On average, each paper was cited 11.5 times.

The number of knowledge diffusion paths emanating from all source nodes to all sink nodes was 3.8 billion plus. It is the purpose of the main path analysis to extract the most significant paths from this huge number of pools. Given the citation network, we calculated the key-route paths based on the methods mentioned in Sect. 3. The whole calculation process was done through a software program developed in-house. The program takes the citation relationship as the major input, turns the relationships into a citation network, determines the SPC values for each citation link, and then searches for the key-route paths.

#### The main knowledge diffusion paths

As mentioned earlier, the key-route approach allows us to examine different levels of details by selecting the number of important links. We start the exploration by generating key-route paths from the top 20, 30, 40, 50, 60, and 70 links with the highest SPC values and find that a divergent-convergent pattern starts to surface visually at 30 links (as shown in Fig. 3). The pattern prevails persistently up to 60 links but then is blurred by other details at 70 links. We therefore determine to analyze the key-route main paths based on the top 60 links. Figure 4 presents this result. In the figures, arrows point from the cited papers to the citing papers. Links with larger SPC values are expressed by the thicker line. The label for each paper begins with the last name of the first author, continues with the first initials of the co-authors (in capital letters), and ends with the publishing year. We draw the key-route main paths in a hexagon-like shape, although they could also be drawn in an arc shape similar to vibrating string clamped at both ends. The reason is that intuitively we think straight lines extending out of and pointing into a node can better convey the idea of divergence and convergence than arcs. The list of the top 60 links with the highest SPC values is presented in Table 1 of Appendix.

Two full cycles of the divergent-convergent pattern are clearly observed in the knowledge diffusion paths in Fig. 3. The third cycle is rather weak and not easy to judge its status. The first cycle starts from PadhiNG1997 (Padhi et al. 1997), who initiated the first idea on the positive-electrode materials of LFP batteries. The second cycle begins with YamadaKSK2005 (Yamada et al. 2005). This study confirmed the possibility of increasing the electronic conductivity of a LFP battery through phase control.

It is noted that one knowledge path (YamadaYTSK2005 to SrinivasanN2006) does not merge to YamadaKSK2005; instead, it crosses through this merging point. There will be more of this type of exception when one zooms in further to examine the knowledge diffusion paths. But this does not change our view of the evolution, as the main structure still prevails.

GibotCLLCHTM2008 (Gibot et al. 2008) begins the third cycle, which shows that the nanoparticle form of LiFePO<sub>4</sub> can be beneficial to the electronic conductivity of a LFP battery.

In the following discussions, we elaborate on the key-route main path drawn on the top 60 links. The discussions focus on the correspondence between the pattern of knowledge diffusion paths and technology development stages. Table 2 in Appendix lists the details of the 56 papers shown on these knowledge diffusion paths. Two citation counts are reported in the table for each paper. "Citations total" is the citation count as indicated in the WOS database. "Citations within dataset" specifies the citation count in the citation network and counts only references from articles in the dataset. Since the articles that refer to articles in the dataset may not themselves be in the dataset, former counts are always equal to or greater than those of the latter.

## First stage: building the fundamentals of LFP battery technology

LiFePO<sub>4</sub> is the chemical formula for LFP. The association of this chemical compound with lithium ion batteries is attributed to Professor John Goodenough's research group at the University of Texas, Austin. Their groundbreaking paper PadhiNG1997 (Padhi et al. 1997) suggested that LiFePO<sub>4</sub> is an excellent candidate for the cathode materials for rechargeable lithium batteries. In the same year, PadhiNMOG1997 (Padhi et al. 1997) reported that,



**Fig. 3** The key-route main paths of LFP battery technology based on the top 30 links. The thickness of each citation link is drawn in proportion to its SPC value. The SPC values of the links in this graph range from  $3.75 \times 10^7$  to  $153.36 \times 10^7$ 



**Fig. 4** The key-route main paths of LFP battery technology based on the top 60 links. The thickness of each citation link is drawn in proportion to its SPC value. The SPC values of the links in this graph range from  $2.76 \times 10^7$  to  $153.36 \times 10^7$ 

among the four iron phosphates (LiFePO<sub>4</sub>,  $Li_3Fe_2(PO_4)_3$ ,  $LiFeP_2O_7$ , and  $Fe_4(P_2O_7)_3$ ), LiFePO<sub>4</sub> exhibits the best electrochemical characteristic.

These two studies (Padhi et al. 1997a, b) established the foundation for the LFP battery technology. All the following works extended, modified, and explored further the ideas of these two studies.

When LiFePO<sub>4</sub> is operating as a cathode material, lithium ions are extracted from LiFePO<sub>4</sub> during the charge. The extraction produces  $FePO_4$ , which bears similar structures

as LiFePO<sub>4</sub>, but with a smaller volume. During discharge, lithium ions are inserted back into FePO<sub>4</sub> and reproduce LiFePO<sub>4</sub>. The charge and discharge process causes a transition between the phases of the delithiated LiFePO<sub>4</sub> and the lithiated FePO<sub>4</sub>. Following PadhiNG1997 and PadhiNMOG1997, AnderssonKHT2000 (Andersson et al. 2000) suggested the existence of the phase interface between the two phases. AnderssonTKH2000 (Andersson et al. 2000) further indicated that the phase interface is the cause of the low capacity for the LFP battery. As we will see later, this two-phase reaction system became one of the central themes for LFP battery technology.

Two streams of studies continued. One stream focused on the synthesis of LiFePO<sub>4</sub> in an effort to determine better ways to produce the compound, especially those with the potential to increase the electronic conductivity of LiFePO<sub>4</sub>. The papers in this stream include YamadaCH2001 (Yamada et al. 2001), YangZW2001 (Yang et al. 2002), ChungBC2002 (Chung et al. 2002), BarkerSS2003 (Barker et al. 2003), and YamadaYTSK2005 (Yamada et al. 2005).

The other stream of studies explored the materials of a similar structure, such as LiMnPO<sub>4</sub>, whereby YamadaC2001 (Yamada and Chung 2001), YamadaKL2001b (Yamada et al. 2001), TuckerDRFRC2002 (Tucker et al. 2002), TangH2003 (Tang and Holzwarth 2003), YamadaHCKHLN2003 (Yamada et al. 2003), DelacourtWRMM2004 (Delacourt et al. 2004), and YonemuraYTSK2004 (Yonemura et al. 2004) are the major works. The results of the second stream of exploration have led to a consensus that LiFePO<sub>4</sub> is a better material than other similar materials in terms of electrochemical performance and structural stability. At this time, the exploration of similar materials was practically completed and the basic synthesis methods of LiFePO<sub>4</sub> were established. This pushed the scientists to shift their endeavors to the study of the two-phase system. The shift was inspired by a series of works, specifically those of Professor Atsuo Yamada's group at the Tokyo University of Technology.

## Second stage: exploring LiFePO<sub>4</sub>/FePO<sub>4</sub> two-phase reaction system

By 2005, a co-existence region of the delithiated LiFePO<sub>4</sub> and the lithiated FePO<sub>4</sub> was well established. The two-phase region's low electron mobility is one of the main causes of low conductivity in the LFP battery. YamadaKSK2005 (Yamada et al. 2005) was the first to experimentally confirm the existence of narrow single-phase regions outside the two-phase co-existence region at room temperature, in contrast to DelacourtPTM2005 (Delacourt et al. 2005), who found a similar region at a very high temperature (450 °C). As was known then, the single-phase LiFePO<sub>4</sub> has a higher electronic conductivity. Consequently, scientists largely expanded their research into the details of the two-phase reaction system. These studies include MaxischZC2006 (Maxisch et al. 2006), DelacourtPLM2006 (Delacourt et al. 2006), LaffontDGWKM2006 (Laffont et al. 2006), SrinivasanN2006 (Srinivasan and Newman 2006), YamadaTKSKIYK2006 (Yamada et al. 2006), YamadaKNSKYNK2006 (Yamada et al. 2006), EllisPRN2006 (Ellis et al. 2006), and ChenSR2007 (Chen et al. 2007). At the end of this stage, FisherI2008 (Fisher and Islam 2008) discussed the two-phase transition behavior on an atomic scale. This led to another shift in research direction which deals with the issues on nanoscale LiFePO<sub>4</sub>.

## Third stage: dealing with the issues on nanoscale LiFePO<sub>4</sub>

A step forward in 2008 triggered a new development in LFP battery technology. GibotCLLCHTM2008 (Gibot et al. 2008) reported that it is possible to "drive the well-established two-phase room-temperature insertion process in LiFePO<sub>4</sub> electrodes into a

single-phase one by modifying the material's particle size and ion ordering" (Gibot et al. 2008). In other words, electrodes made of  $LiFePO_4$  nanoparticles exhibited single-phase behavior. This discovery set a new research direction toward issues on the two-phase reaction and synthesis of LiFePO<sub>4</sub> on a nanoscale. This stream of studies include RamanaMGJZ2009 (Ramana et al. 2009), AxmannSWMGJ2009 (Axmann et al. 2009); Adams 2010 (Adams 2010), and AdamsR2011a (Adams and Rao 2011), while at the same time research on the two-phase reaction system continues. These include KobayashiNPKYIY2009 (Kobayashi et al. 2009), MatsuiNKTY2010 (Matsui et al. 2010), and a series of studies by Safari and Delacourt (2011a, b, c).

#### **Discussion and conclusions**

Technological change is characterized by cyclical and reciprocal links between divergence and convergence. The divergence starts with a breakthrough innovation which leads to a chaotic period when a variety of technologies compete for their influence. The convergence begins as these competing technologies gradually combine or fuse together into dominant designs. Technological change as successive cycles is easier to intuit than to define and measure. To address this challenge, we extend recent work on defining technological change in terms of knowledge flows to visualize change as cycles of knowledge diffusion paths.

In particular, the study of LFP battery technology allows us to illustrate the usefulness of the key-route main path analysis on technological changes and cycles, while at the same time, producing insightful case descriptions. We have illustrated the LFP battery technology that evolved in a cyclical pattern of repetitive divergence-convergence links. Two cycles of technological development are identified. The first cycle of evolution started in 1997, when the idea of using LiFePO<sub>4</sub> as a cathode material was first proposed. The second cycle of development began in 2005, when the existence of the LiFePO<sub>4</sub> solid solution was discovered, triggering a new wave of research on the two-phase reaction system. In 2008, a new research direction emerged which focuses on the issues of the two-phase reaction and synthesis of LiFePO<sub>4</sub> on a nanoscale. As of March 2012, the third stage, or cycle of development, has yet to see its termination.

In order to verify the divergence-convergence pattern observed in the topology of the key-route main paths, we consulted domain experts, who are local and within our access, on the results of the analysis. There is a general consensus on the breakthrough points of the first cycle and the second cycle. The domain experts, however, do not completely agree on whether the third cycle has actually begun. The disagreement is somewhat expected because both the methodology and experts are observing something that is too close in time on which to make proper judgment.

It is important to recognize that our analysis is subject to several limitations. First, there exists time span bias for this particular LFP battery technology analysis. The LFP battery technology has only been developed for 15 years. In such a short time span, we observe two small scale paradigmatic shifts framed under the concept of divergence-convergence links. In the longer term, as the efforts in increasing the battery efficiency continue, new and bigger discontinuities may arrive on the scene and make the changes we observe at this moment ripple in a long wave. Thus, the key milestones in the divergent-convergent pattern of a particular technology may change as it evolves along, but the pattern should prevail.

Second, the number of top citation links selected in drawing key-route main paths is an arbitrary number that needs to be selected with some care. On one hand, a small number

will reduce the key-routes main paths to too few paths, such that no divergent-convergent pattern can be observed. On the other hand, a very large number may add paths that include less significant citation links, which blur the governing structure of the knowledge diffusion paths. To the extreme, the key-route main paths will become the original citation networks, which defeat the purpose of applying the main path analysis. We have observed that in the LFP battery technology case, top citation links between 30 and 60 exhibit relatively clear divergent-convergent patterns. As the number increases, the pattern is gradually blurred by less significant citation links.

Third, the analysis is unable to predict explicitly the turning point of the technological cycles, nor is it able to identify the precedence and subsequence of the technology evolutionary path. Similarly, it is difficult for the analysis to examine the pattern of technology development in multidisciplinary studies. The purpose of the study is to show the effectiveness of using an innovative approach (the key-route main analysis) to identify the cyclical patterns of technology development in a clearly defined discipline.

Despite the limitations, this study provides important implications for research and contributes to at least two key issues in the study of technological change. First, our study contributes to our understanding of technological change as a succession of the technology cycle. We identify each cycle beginning with a technological discontinuity that inaugurates an era of technology divergence and design competition. The technological divergence culminates in the appearance of a dominant design that is elaborated upon through the process of the technology convergence, until a new technology displaces its predecessor. The concept of the divergent-convergent pattern enriches the technological change literature with a topological view of evolution.

The second issue to which our study contributes is the nature of knowledge flows. We measure knowledge flows in LFP battery technology using paper citation data and employ a novel key-route method to detect the main diffusion paths of knowledge flow from the earlier/cited paper to the later/citing paper. Paper citations reflect the actual flow of ideas for new technology that corresponds to intellectual influence and conscious knowledge spillover. Because a generational and cyclical shift brings about major changes at the knowledge level, identification of the path with the highest overall connectivity in paper citations helps reveal the critical backbone of knowledge flow within the formation of each different technology regime and cycle.

In the future, several additional aspects can be further explored to enhance the observation of the current study. First, given that science and technology literature has long depicted that technology evolves in a cyclical pattern, we speculate that other technologies or technology systems could have similar cyclical patterns as we have demonstrated for LFP battery technology. It will be interesting to observe if other technologies or technology systems bear the similar topological pattern. Second, technological changes should also be reflected in the variation of keyword usages in time. The yearly distribution of keyword frequencies can be examined in parallel with the main path analysis results and may provide supplementary supporting evidence for technological changes.

### Appendix

See Appendix Tables 1 and 2.

Table 1 List of the top 60 links			
Ranks	SPC values	From	To
1	1533591198	PadhiNG1997 (952)	PadhiNMOG1997 (225)
2	694399380	YamadaYTSK2005 (20)	YamadaKSK2005 (63)
3	672237250	AnderssonKHT2000 (218)	YamadaCH2001 (499)
4	630020844	YonemuraYTSK2004 (58)	YamadaYTSK2005 (20)
5	602182944	YamadaHCKHLN2003 (77)	Yamada2003 (8)
6	557348333	PadhiNG1997 (952)	AnderssonKHT2000 (218)
7	557348333	PadhiNMOG1997 (225)	AnderssonKHT2000 (218)
8	536681032	Fisher12008 (28)	GibotCLLCHTM2008 (63)
6	513778750	YamadaTKSKIYK2006 (25)	YamadaKNSKYNK2006 (98)
10	458356580	DelacourtPTM2005 (109)	YamadaKSK2005 (63)
11	432324750	SrinivasanN2006 (43)	YamadaKNSKYNK2006 (98)
12	429706934	PadhiNG1997 (952)	AnderssonTKH2000 (92)
13	429706934	PadhiNMOG1997 (225)	AnderssonTKH2000 (92)
14	412593180	YonemuraYTSK2004 (58)	YamadaKSK2005 (63)
15	388725660	YamadaKSK2005 (63)	SrinivasanN2006 (43)
16	381066820	YangSZW2002 (129)	ChungBC2002 (558)
17	374730480	YamadaKSK2005 (63)	MaxischZC2006 (61)
18	365805635	Yamada2003 (8)	YonemuraYTSK2004 (58)
19	365805635	Yamada2003 (8)	YonemuraYTSK2004 (58)
20	355261990	DelacourtWRMM2004 (8)	DelacourtPTM2005 (109)
21	339683554	YamadaKL2001b (62)	TuckerDRFRC2002 (15)
22	336118625	PadhiNMOG1997 (225)	YamadaCH2001 (499)
23	336118625	PadhiNG1997 (952)	YamadaCH2001 (499)
24	299693430	YamadaKSK2005 (63)	YamadaTKSKIYK2006 (25)
25	281246180	MaxischZC2006 (61)	DelacourtPLM2006 (123)
26	258836224	TuckerDRFRC2002 (15)	BarkerSS2003 (119)

Ranks     SPC values     From     To       27     24891260     YamadaKSK2005 (63)     Yam       28     249342609     YamadaKSK2005 (63)     Yam       28     249342609     YamadaKSKYYK2005 (15)     Tuck       29     24940052     YamadaKSKYYK2006 (98)     Huta       20     24940052     Anderssorf/KH2006 (92)     Huta       31     228881522     Anderssorf/KH2006 (92)     Huta       32     22393175     TuckerDRFCR002 (20)     Huta       33     212393175     TuckerDRFCR002 (20)     Yam       34     203494464     YamadaCH2001 (499)     Yam       37     203497464     YamadaCH2001 (499)     Yam       38     2044440     YamadaCH2001 (499)     Yam       39     19885562     YamadaCH2001 (499)     Yam       41     191813421     YamadaCH2001 (499)     Yam       39     19383562     YamadaCH2001 (499)     Yam       40     19572772     YamadaCH2001 (499)     Yam       41     191981342     YamadaC	Table 1 continued			
27     24,89,12,60     YamadaKSK2005 (63)     Yam       28     248,342,80     YamadaKSK2005 (53)     Yam       29     248,342,80     YamadaKSK2005 (53)     Yam       29     244,965,58     TuckerDRFRC2001 (55)     Yam       31     228,581,52     AnadaKNSKYNK2006 (98)     Hulu       32     224,340,52     AnaderssonTKH2000 (21)     Hulu       33     285,5873     AnderssonTKH2000 (22)     Hulu       34     2046443     YamadaKNSKYNK2006 (98)     Yam       35     2046444     YamadaKD01 (499)     Yam       36     2046444     YamadaKD01 (499)     Yam       37     2044460     YamadaKD01 (499)     Yam       38     20444460     YamadaKD01 (499)     Yam       39     203947464     YamadaKD01 (499)     Yam       36     2044460     YamadaKD01 (499)     Yam       37     20444400     YamadaKD01 (499)     Yam       38     20444460     YamadaKD1200 (499)     Yam       39     1981427     YamadaKD1200	Ranks	SPC values	From	To
28     24834280     YamadaC2001 (85)     Yam       29     2443628     TuckerDRFRC2002 (15)     Tuck       31     224381522     AnderssonTKH2000 (92)     Hun       32     224381522     AnderssonTKH2000 (22)     Hun       33     22858152     Morgan/C2004 (163)     Hende       34     22848425     Morgan/C2004 (163)     Hende       35     22488442     Yamada/C2001 (45)     Hende       36     212393175     TuckerDRFRE2007 (15)     Hende       37     207644030     ChenSR2007 (15)     Panta       38     203947461     Yamada/C12001 (499)     Yam       38     2044050     Yamada/C12001 (499)     Yam       39     203447460     Yamada/C12001 (499)     Yam       41     19981342     Yamada/C12003 (7)     Yam       42     19981342     Yamada/CN06 (98)     Yam       43     191981342     Yamada/CN003 (7)     Yam       44     1572727     Yamada/CN003 (7)     Yam       45     1572475     Yamada/CN00	27	254891250	Y amadaKSK2005 (63)	YamadaKNSKYNK2006 (98)
29     23495628     TuckerDRFRC2002 (15)     Tuck       30     23434052     YamadaKNSKYNK2006 (98)     Filis       31     22881522     AnderssorTKH2000 (92)     Hand       32     224884425     MorganVC2004 (163)     Hand       33     2124884425     MorganVC2004 (163)     Hand       34     20365873     HateECDSC004 (231)     Hand       35     207644030     ChenSR2007 (15)     YamadaCH2001 (499)     YamadaCH2001 (499)       36     20364746     YamadaCH2001 (499)     YamadaCH2001 (499)     YamadaCH2001 (499)     YamadaCH2001 (499)       37     203647460     YamadaCH2001 (499)     YamadaCH2001 (499)     YamadaCH2001 (499)     YamadaCH2001 (499)     YamadaCH201 (490)     YamadaCH201 (490)     YamadaCH201 (490)     YamadaCH201 (490)     YamadaCH201 (490)     Y	28	248342809	YamadaC2001 (85)	YamadaKL2001b (62)
30 23434052 YamadaKNSKYNK2006 (98) Ellis   31 238581522 AnderssonTKH2000 (92) Hun   32 228581522 MorganVC2004 (163) Hede   33 212393175 TuckerDRFCR2002 (20) Yam   34 203653873 HerleECN2004 (231) Delta   35 20464403 ChenSEO07 (15) Yam   36 203947461 YamadaCH2001 (499) Yam   37 20364730 ChenSEO03 (8) Yam   38 20444460 YamadaCH2001 (499) Yam   39 203947461 YamadaCH2001 (499) Yam   36 203447460 YamadaCH2001 (499) Yam   37 203447460 YamadaCH2001 (499) Yam   38 20444400 YamadaCH2001 (499) Yam   39 19885562 YamadaCH2001 (499) Yam   40 19752772 YamadaCH2001 (499) Yam   41 19881342 YamadaCH2011 (499) Yam   42 181242176 YamadaCH2KHNS2003 (77) Yam   43 1175128 YamadaCH2KHNS2003 (77) Yam   44 1751288 YamadaCH2KHNS2003 (77) Yam   45 1751298 YamadaCH2KHNS2003 (77)	29	234956288	TuckerDRFRC2002 (15)	TuckerDRFCR2002 (20)
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33   212393175   TuckerDRFCR2002 (20)   Yam     34   208653873   HerleECN2004 (231)   Fishe     35   207644030   ChenSR2007 (15)   Fishe     36   20468644   YamadaCH2001 (499)   Yam     37   203947464   YamadaCH2001 (499)   Yam     38   20044460   YamadaCH2001 (499)   Yam     39   199885562   MaierA2008 (9)   Gibo     40   19572772   YamadaCH2001 (499)   Yam     41   19981342   YamadaCH2001 (499)   Yam     42   19981342   YamadaCH2003 (8)   Gibo     43   19981342   YamadaKNSKYNK2006 (98)   Laffe     44   1512988   YamadaKNSKYNK2006 (98)   Laffe     45   17612988   YamadaKNSKYNK2006 (98)   Delaa     46   1770   TucketDRFRC2002 (15)   Yam     47   16129293   YamadaKNSKYNK2006 (98)   Delaa     48   175349508   YamadaKNSKYNK2006 (98)   Pelaa     49   17612988   YamadaKNSKYNK2006 (98)   Pelaa     46   177747420	32	224884425	MorganVC2004 (163)	HerleECN2004 (231)
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39   19985562   MaierA2008 (9)   Gibo     40   195722772   YamadaKNSKYNK2006 (98)   Laffe     41   191981342   YamadaKNSKYNK2006 (98)   Laffe     42   191981342   YamadaKNSKYNK2006 (98)   Amir     43   184520512   YamadaKNSKYNK2006 (98)   Amir     44   17612988   YamadaHCKHLN2003 (77)   Yam     45   TuckerDRFRC2002 (15)   Yam   Yam     46   17612988   YamadaHCKHLN2003 (77)   Delax     47   175349508   YamadaHCKHLN2003 (77)   Yam     48   174744720   GibotCLLCHTM2008 (63)   Yam     49   174744720   GibotCLLCHTM2008 (63)   Yam     41   169581844   YamadaKNSKYNK2006 (98)   Meet     49   169581844   YamadaKNSKYNK2006 (98)   Yam     49   169780750   TangH2003 (29)   Yam     49   167800750   TangH2003 (29)   Yam     40   164703786   ChungBC2002 (558)   Yam     50   164112158   BarkerSS2003 (119)   Yam     51   16411203	38	200444460	Yamada2003 (8)	YamadaYTSK2005 (20)
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41   191981342   PadhiNG1997 (952)   Amir     42   184520512   YamadaHCKHLN2003 (77)   Delaa     43   181242176   TuckerDRFRC2002 (15)   Yam     44   17612988   YamdaHCKHLN2003 (77)   Delaa     45   17612988   Yam2W2001 (188)   Yam     46   175349508   GibotCLLCHTM2008 (63)   Yam     47   169581844   YamadaKNSKYNK2006 (98)   Ram     48   174744720   TangH2003 (29)   Yone     49   167800750   TangH2003 (29)   Yone     50   164703786   ZaghibMGGI2007 (41)   Fishe     51   163084125   BarkerSS2003 (119)   Morg     53   16112158   BarkerSS2003 (119)   Hetle	40	195722772	YamadaKNSKYNK2006 (98)	LaffontDGWKMT2006 (74)
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43   181242176   TuckerDRFRC2002 (15)   Yam     44   17612988   YamadaHCKHLN2003 (77)   Dela     45   175349508   YamZW2001 (188)   Dela     46   174744720   GibotCLLCHTM2008 (63)   Ram     47   169581844   YamadaKNSKYNK2006 (98)   Meet     48   169581844   YamadaKNSKYNK2006 (98)   Meet     49   167800750   TangH2003 (29)   Yone     50   164703786   ZaghibMGGI2007 (41)   Fishe     51   163084125   BarkerSS2003 (119)   Morg     52   161705430   VanadaKNSKY3005 (63)   Morg	42	184520512	YamadaHCKHLN2003 (77)	DelacourtWRMM2004 (8)
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Ranks	SPC values	From	To
53	157717288	GaberscekDJ2007a (48)	GibotCLLCHTM2008 (63)
54	153367596	HerleECN2004 (231)	YamadaYTSK2005 (20)
55	152787468	YamadaYTSK2005 (20)	SrinivasanN2006 (43)
56	145965892	YamadaKNSKYNK2006 (98)	MeethongHSCC2007 (57)
57	141240100	YamadaCH2001 (499)	YamadaKL2001a (54)
58	140097060	GibotCLLCHTM2008 (63)	KobayashiNPKYIY2009 (29)
59	138667138	AnderssonTKH2000 (92)	ProsiniZP2001 (148)
60	137555847	EllisHRNDPR2007 (18)	RhoNPR2007 (39)
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-The label for each article begins with the last name of the first author, continues with the initials of the co-authors, and ends with its publishing year. The numbers in parentheses indicate the times each article is cited by others in the dataset

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Table 2	Label

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Label	Authors	Title	Citations total	Citations within dataset
PadhiNG 1997	Padhi, AK; Nanjundaswamy, KS; Goodenough, JB	Phospho-olivines as positive-electrode materials for rechargeable lithium batteries	1895	952
ChungBC2002	Chung, SY; Bloking, JT; Chiang, YM	Electronically conductive phospho-olivines as lithium storage electrodes	1001	558
YamadaCH2001	Yamada, A; Chung, SC; Hinokuma, K	Optimized LiFePO <sub>4</sub> for lithium battery cathodes	776	499
HuangYN2001	Huang, H; Yin, SC; Nazar, LF	Approaching theoretical capacity of LiFePO4 at room temperature at high rates	482	461
PadhiNMOG 1997	Padhi, AK; Nanjundaswamy, KS; Masquelier, C; Okada, S; Goodenough, JB	Effect of structure on the Fe3 +/Fe2 + redox couple in iron phosphates	451	225
HerleECN2004	Herle, PS; Ellis, B; Coombs, N; Nazar, LF	Nano-network electronic conduction in iron and nickel olivine phosphates	388	231
AnderssonKHT2000	Andersson, AS; Kalska, B; Haggstrom, L; Thomas, JO	Lithium extraction/insertion in LiFePO <sub>4</sub> : an X-ray diffraction and Mossbauer spectroscopy study	311	218
YangZW2001	Yang, SF; Zavalij, PY; Whittingham, MS	Hydrothermal synthesis of LFP cathodes	287	188
MorganVC2004	Morgan, D; Van der Ven, A; Ceder, G	Li conductivity in LixMPO4 ( $M = Mn$ , Fe, Co, Ni) olivine materials	277	163
ProsiniZP2001	Prosini, PP; Zane, D; Pasquali, M	Improved electrochemical performance of a LiFePO4-based composite cathode	223	148
Amine YY2000	Amine, K; Yasuda, H; Yamachi, M	Olivine LiCoPO4 as 4.8 V electrode material for lithium batteries	215	129
DelacourtPLM2006	Delacourt, C; Poizot, P; Levasseur, S; Masquelier, C	Size effects on carbon-free LiFePO4 powders	199	123
YangSZW 2002	Yang, SF; Song, YN; Zavalij, PY; Whittingham, MS	Reactivity, stability and electrochemical behavior of LFPs	198	129
DelacourtPTM2005	Delacourt, C; Poizot, P; Tarascon, JM; Masquelier, C	The existence of a temperature-driven solid solution in LixFePO4 for $0 <= x <= 1$	170	109
YamadaC2001	Yamada, A; Chung, SC	Crystal chemistry of the olivine-type Li(MnyFe1-y)PO4 and (MnyFe1-y)PO4 as possible 4 V cathode materials for lithium batteries	166	85

Table 2 continued				
Label	Authors	Title	Citations total	Citations within dataset
AnderssonTKH2000	Andersson, AS; Thomas, JO; Kalska, B; Haggstrom, L	Thermal stability of LiFePO4-based cathodes	152	86
YamadaKNSNKYNK2006	Yamada, A; Koizumi, H; Nishimura, SI; Sonoyama, N; Kanno, R; Yonemura, M; Nakamura, T; Kobayashi, Y	Room-temperature miscibility gap in LixFePO4	152	92
ChenSR2006	Chen, GY; Song, XY; Richardson, TJ	Electron microscopy study of the LiFePO4 to FePO4 phase transition	151	76
YamadaHCKHLN2003	Yamada, A; Hosoya, M; Chung, SC; Kudo, Y; Hinokuma, K; Liu, KY; Nishi, Y	Olivine-type cathodes achievements and problems	147	77
GibotCLLCHTM2008	Gibot, P; Casas-Cabanas, M; Laffont, L; Levasseur, S; Carlach, P; Hamelet, S; Tarascon, JM; Masquelier, C	Room-temperature single-phase Li insertion/extraction in nanoscale LixFePO4	130	63
MeethongHSCC2007	Meethong, N; Huang, HYS; Speakman, SA; Carter, WC; Chiang, YM	Strain accommodation during phase transformations in olivine-based cathodes as a materials selection criterion for high-power rechargeable batteries	124	57
YonemuraYTSK2004	Yonemura, M; Yamada, A; Takei, Y; Sonoyama, N; Kanno, R	Comparative kinetic study of olivine LixMPO4 ( $M = Fe$ , $Mn$ )	120	58
LaffontDGWKMT2006	Laffont, L; Delacourt, C; Gibot, P; Wu, MY; Kooyman, P; Masquelier, C; Tarascon, JM	Study of the LiFePO4/FePO4 two-phase system by high-resolution electron energy loss spectroscopy	119	74
BakerSS2003	Barker, J; Saidi, MY; Swoyer, JL	Lithium iron(II) phospho-olivines prepared by a novel carbothermal reduction method	115	119
MeethongHCC2007	Meethong, N; Huang, HYS; Carter, WC; Chiang, YM	Size-dependent lithium miscibility gap in nanoscale Li1-xFePO4	109	66
YamadaKL2001b	Yamada, A; Kudo, Y; Liu, KY	Phase diagram of Li- $x(MnyFe1-y)PO4$ (0 <= x, y <= 1)	106	62
YamadaKL2001a	Yamada, A; Kudo, Y; Liu, KY	Reaction mechanism of the olivine-type Li-x(Mn0.6Fe0.4)PO4 $(0 \le x \le 1)$	98	54
GaberscekDJ2007a	Gaberscek, M; Dominko, R; Jamnik, J	Is small particle size more important than carbon coating? An example study on LiFePO4 cathodes	93	48

Table 2 continued				
Label	Authors	Title	Citations total	Citations within dataset
YamadaKSK2005	Yamada, A; Koizumi, H; Sonoyama, N; Kanno, R	Phase change in LixFePO4	79	63
MaxischZC2006	Maxisch, T; Zhou, F; Ceder, G	<i>Ab initio</i> study of the migration of small polarons in olivine LixFePO4 and their association with lithium ions and vacancies	79	61
SrinivasanN2006	Srinivasan, V; Newman, J	Existence of path-dependence in the LiFePO <sub>4</sub> electrode	62	43
EllisPRN2006	Ellis, B; Perry, LK; Ryan, DH; Nazar, LF	Small polaron hopping in LixFePO4 solid solutions: Coupled lithium- ion and electron mobility	62	39
RhoNPR2007	Rho, YH; Nazar, LF; Perry, L; Ryan, D	Surface chemistry of LiFePO4 studied by mossbauer and X-ray photoelectron spectroscopy and its effect on electrochemical properties	62	39
ZaghibMGGJ2007	Zaghib, K; Mauger, A; Goodenough, JB; Gendron, F; Julien, CM	Electronic, optical, and magnetic properties of LiFePO4: Small magnetic polaron effects	59	41
Fisher12008	Fisher, CAJ; Islam, MS	Surface structures and crystal morphologies of LiFePO <sub>4</sub> : Relevance to electrochemical behavior	53	28
KobayashiNPKYIY2009	Kobayashi, G; Nishimura, SI; Park, MS; Kanno, R; Yashima, M; Ida, T; Yamada, A	Isolation of Solid Solution Phases in Size-Controlled $Li(x)FePO(4)$ at Room Temperature	52	29
TuckerDRFCR2002	Tucker, MC; Doeff, MM; Richardson, TJ; Finones, R; Cairns, EJ; Reimer, JA	Hyperfine fields at the Li site in LiFePO <sub>4</sub> -type olivine materials for lithium rechargeable batteries: A Li-7 MAS NMR and SQUID study	48	29
TangH2003	Tang, P; Holzwarth, NAW	Electronic structure of FePO4, LiFePO4, and related materials	48	20
YamadaTKSKIYK2006	Yamada, A; Takei, Y; Koizumi, H; Sonoyama, N; Kanno, R; Itoh, K; Yonemura, M; Kamiyama, T	Electrochemical, magnetic, and structural investigation of the Li- x(MnyFe1-y)PO4 olivine phases	43	25
RamanaMGJZ2009	Ramana, CV; Mauger, A; Gendron, F; Julien, CM; Zaghib, K	Study of the Li-insertion/extraction process in LiFePO <sub>4</sub> /FePO <sub>4</sub>	41	22
EllisHRNDPR2007	Ellis, B; Herle, PS; Rho, YH; Nazar, LF; Dunlap, R; Perry, LK; Ryan, DH	Nanostructured materials for lithium-ion batteries: Surface conductivity vs. bulk ion/electron transport	39	18
TuckerDRFRC2002	Tucker, MC; Doeff, MM; Richardson, TJ; Finones, R; Reimer, JA; Cairns, EJ	Li-7 and P-31 magic angle spinning nuclear magnetic resonance of LiFePO <sub>4</sub> -type materials	35	15

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Label	Authors	Title	Citations total	Citations within dataset
Yamada YTSK2005	Y amada, A; Yonemura, M; Takei, Y; Sonoyama, N; Kanno, R	Fast charging LiFePO <sub>4</sub>	30	20
ChenSR2007	Chen, GY; Song, XY; Richardson, TJ	Metastable solid-solution phases in the LiFePO4/FePO4 system	28	15
DelacourtWRMM2004	Delacourt, C; Wurm, C; Reale, P; Morcrette, M; Masquelier, C	Low temperature preparation of optimized phosphates for Li-battery applications	24	8
MaierA2008	Maier, J; Amin, R	Defect chemistry of LiFePO4	18	6
AxmannSWMGJ2009	Axmann, P; Stinner, C; Wohlfahrt-Mehrens, M; Mauger, A; Gendron, F; Julien, CM	Nonstoichiometric LiFePO4: Defects and related properties	17	10
Adams2010	Adams, S	Lithium ion pathways in LiFePO(4) and related olivines	8	4
SafariD2011a	Safari, M; Delacourt, C	Mathematical Modeling of LFP Electrode: Galvanostatic Charge/ Discharge and Path Dependence	٢	9
Yamada2003	Yamada, A	Materials of lithium battery for the next generation. 3. Iron-based cathodes matching industrial & environmental requirements; Towards inexpensive yet reliable large-scale lithium secondary batteries	Q	×
AdamsR2011a	Adams, S; Rao, RP	Simulated defect and interface engineering for high power Li electrode materials	4	ę
SafariD2011b	Safari, M; Delacourt, C	Modeling of a Commercial Graphite/LiFePO(4) Cell	4	2
MatsuiNKTY2010	Matsui, H; Nakamura, T; Kobayashi, Y; Tabuchi, M; Yamada, Y	Open-circuit voltage study on LiFePO(4) olivine cathode	б	б
DelacourtAT2011	Delacourt, C; Ati, M; Tarascon, JM	Measurement of Lithium Diffusion Coefficient in Li(y)FeSO(4)F	2	1
SafariD2011c	Safari, M; Delacourt, C	Aging of a Commercial Graphite/LiFePO(4) Cell	2	1
FarkhondehD2012	Farkhondeh, M; Delacourt, C	Mathematical Modeling of Commercial LiFePO4 Electrodes Based on Variable Solid-State Diffusivity	0	0

117

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