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Perovskite and related oxide based electrodes for water splitting

Ning Han [a], Marco Race [b,*], Wei Zhang [a], Raffaele Marotta [c], Chi Zhang [d], Awais Bokhari [e,**], Jiří Jaromír Klemeš [e]

[a] Department of Materials Engineering, KU Leuven, Leuven, 3001, Belgium
[b] Department of Civil and Mechanical Engineering, University of Cassino and Southern Lazio, Via di Biasio 43, 03043, Cassino, Italy
[c] DICMAPI, Università di Napoli Federico II, p.le V. Tecchio 80, 80125, Napoli, Italy
[d] School of Applied Physics and Materials, Wuyi University, Jiangmen, 529020, China
[e] Sustainable Process Integration Laboratory - SPIL, NETME Centre, Faculty of Mechanical Engineering, Brno University of Technology - VUT Brno, Technická 2896/2, 616 69, Brno, Czech Republic

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ABSTRACT

Water splitting for renewable energy technologies necessitates the creation of low-cost, highly active, and stable electrocatalysts for the hydrogen evolution reaction (HER) and oxygen evolution reaction (OER), which remains a challenging issue. Oxide-based catalysts, especially perovskites (ABO_3), ruddlesden–popper perovskite (A_2BO_4) and pyrochlore-type oxides ($A_2B_2O_7$), have attracted gotten much interest as electrocatalysts for water splitting because of their distinctive physical, chemical, and electronic properties. In this work, a scientometric analysis is conducted on developing various oxide-based catalysts for water splitting to explore further developments. Up to now, a total of 29,761 publications (between the years 1990–2020) have been obtained from the Web of Science (WoS) website by searching for these written documents on this subject with a variety of linking keywords. The derived documents were subjected to a scientometric study, which included a look at the contributing authors, publications, contributing countries, top citation bursts, and topic categories. Based on the information obtained, in this research area, the publications started to increase dramatically since 2012. In terms of scientific output on oxide-based catalysts for water splitting, China and the United States were discovered to be the world leaders.

1. Introduction

The worldwide increasing energy demand generated by population growth has resulted in the fast use of the fossil fuels (Wang et al., 2013) and serious environmental problems (Kumar et al., 2020). The reduction of the supplies of fossil fuel and the urgent need to reduce emissions of greenhouse gas to tackle climate change have driven us into the more sustainable, renewable (Ekanayake et al., 2020), low-cost (Liu, K. et al., 2020), and safer alternative to the current system of energy generation, such as clean oxygen production (Han et al., 2019) through CO_2 resistance membranes (Zhang et al., 2017), magnesium-sulphur batteries (Razaq et al., 2020), fruit-based batteries (Wang, Z. et al., 2021), solar water splitting cells (Walter et al., 2010), and artificial photosynthesis (Xie et al., 2011). Hydrogen is a possible alternative energy carrier for replacing the conventional fossil fuel grid because of the high mass-energy density and lack of greenhouse gas emissions (Yuan et al., 2021). The availability of cost-effective hydrogen processing technologies is needed for the implementation of such renewable energy schemes (Hwang et al., 2017). The majority of hydrogen production is faced by the expensive and energy-intensive steam reforming of hydrocarbons (He and Li, 2015), which are produced from fossil fuels and release a significant number of pollutants as a result. Despite the technological challenges, electro- (Wang, Y. et al., 2020) or photochemical (Du et al., 2020) dissociation of water is a promising method of producing hydrogen. The ultimate water-splitting reaction consists of two distinct reactions: hydrogen evolution and oxygen evolution reactions (HER and OER) (Z. Huang et al., 2019). The overall reaction in water splitting is displayed in Equation (1). The following are the HER and OER reaction equations in the alkaline electrolyte - Equations (2-3) and acidic electrolyte - Equations (4-5):

$$2H_2O \rightarrow 2H_2 + O_2, \Delta E = -1.23 \text{ V} \quad (1)$$

Basic electrolyte:

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* Corresponding author.
** Corresponding author.
E-mail addresses: marco.race@unicas.it (M. Race), bokhari@fme.vutbr.cz (A. Bokhari).

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$$\text{OER: } 4OH^{-} \rightarrow 2H_2O + O_2 + 4e^{-} \tag{2}$$

$$\text{HER: } 4H_2O + 4e^{-} \rightarrow 2H_2 + 4OH^{-} \tag{3}$$

Acidic electrolyte:

$$\text{OER: } 2H_2O \rightarrow 4H^{+} + O_2 + 4e^{-} \tag{4}$$

$$\text{HER: } 2H^{+} + 2e^{-} \rightarrow H_2 \tag{5}$$

To accelerate HER and OER, the new benchmark electrolyse employs a Pt-based cathode and RuO_2/IrO_2 anode (Han et al., 2018). However, effective electrocatalysts are normally needed for both reactions to proceed smoothly (Liu, P.F. et al., 2020). Until now, noble metal-based materials such as Ir (Wang, H. et al., 2021), Ru (Yu et al., 2019), and Pt (Zhang et al., 2016a,b) have been shown to be the ideal catalysts for the electrochemical water splitting mechanism or water electrolysis. The high cost and shortage of these precious metals, on the other hand, have severely limited their widespread use (Zhang et al., 2020). From the standpoint of commercialisation, it is not just the high cost of noble metal components that causes economic strain (Wang et al., 2017), but also the increased cost incurred due to the difficulties of manufacturing multiple cathode–anode products and potential cross-contaminations (Suntivich et al., 2011b). As a result, much effort has gone into developing efficient and low-cost electrocatalysts for water splitting (You and Sun, 2018). Various earth-abundant non-precious metal electrocatalysts have been evaluated in terms of HER, OER, and overall water splitting efficiency (Charles et al., 2021). The majority of these promising catalysts are perovskite oxide (Xu et al., 2016), Ruddlesden-Popper oxide (Zhu et al., 2020), phosphides (Huo et al., 2019), carbon-based semiconductors (Kumar et al., 2018), metal carbides (Michalsky et al., 2014), metallic carbides (Qiao et al., 2020), carbon-supported single-atom catalysts (Yang et al., 2020), alkoxides (Liu, X. et al., 2020), nitrides (Han et al., 2018), hydroxides (Liu et al., 2019), hybrids (Yu et al., 2021), graphene-based composites (Li et al., 2017), and other compositional engineering designed materials (Sun et al., 2020).

Oxide catalysts emerged as a hot family of electrochemical catalysts (Beall et al., 2021), attracting increasing attention from researchers because of their peculiar physical, chemical (Wei et al., 2017), and electronic properties (Cheng et al., 2018). Any of them exhibit outstanding catalytic operations in several reactions relevant to energy applications. About 30 thousand publications on oxide catalysts for water splitting have been made to-date (from Jan 1990 to Dec 2020, based on ISI Web of Science). The number of documents that have been released strongly emphasises the possible use of oxide-based catalysts for water splitting in conjunction with other conventional fossil fuel energy systems for hydrogen production. These aspects can be attributed to the low cost, high stability and smart flexibility from designing catalysts. Meanwhile, the multiple applications (advanced oxidation (Han et al., 2020), gas separation membrane (Han et al., 2019), solid electrolyte (Han and Yildiz, 2012), metal-air batteries (Suntivich et al., 2011a), as reducing reagent (Iizuka et al., 2011), solid state fuel cells (Yao et al., 2021), as phase transition materials (Chen et al., 2020), oxygen reduction (Stoerzinger et al., 2015), oxygen evolution (Suntivich et al., 2011b), and photocatalytic catalyst (Wang, H. et al., 2020)) have been developed because of their excellent physio-chemical properties (Lee et al., 2016), especially single perovskites (Han et al., 2019), double perovskites (Kim et al., 2014), Ruddlesden–Popper perovskites (Han et al., 2021), and pyrochlore-type oxides (Zeng et al., 2007), which also proves the potential as oxide catalysts for electrochemical (Wang et al., 2020) and photoelectrochemical water splitting (Wang, W. et al., 2020).

The details in similar publications may represent the characteristics and patterns of research on a discipline (Hwang et al., 2017). It is not sufficient scientific, statistical analysis work comprehensively covering oxides for photo-, electro-, and photoelectro-water splitting. With the booming of the research on oxide catalysts for water splitting, it is urgent to summary the existing research using an appropriate means to assess the characteristics and trends of the research on this area.

The present work evaluated the present state of science and technological advancement in the field of oxide-based catalysts, especially perovskites, Ruddlesden–Popper perovskites, and pyrochlore-like oxides for water splitting. The scientometric study and knowledge visualisation analysis performed here seek to analyse the latest emerging developments in this research field, which will aid in identifying vulnerabilities and areas for further improvement to accelerate fundamental understanding and commercialisation of oxide-based catalysts for energy storage systems. An analytical examination of the present state of the art in this field would be highly beneficial in bolstering the theoretical conclusion based on scientific experience, advances achieved, past and existing developments in this field, and finding holes and potentials for possible advancements. The current study offers a thorough scientometric study of attempts worldwide to create oxide-based water-splitting catalysts. The collected data would validate research trends and provide a comprehensive view of scientific progress in this field.

2. Method

The archive was chosen to download the relevant documentation from the ISI Web of Science (WoS) core collection to begin the scientometric research, including indexed papers, conference proceedings records, and other materials. For visualised comparisons, Anaconda Prompt and ScientoPy (v.2.0.3.7z) with Python were used, and network visualisation was used for result treatment and presentation. The keywords: “TS=(*oxide* or *Ruddlesden-Popper* or *RP* or RP_{214} or A_2BO_4 or AB_2O_4 or *Perovskite* or *double-perovskite* or **perovskite** or $A_2B_2O_6$ or ABO_3 or $A_2B_2O_{5.5}$ or *Pyrochlore* or *Pyroch** or $A_2B_2O_7$) and (*OER* or *HER* or *water splitting* or *oxygen evolution* or *hydrogen evolution*), for the overall oxide-based catalysts, the advance quest mode of the WoS core array was used (Table 1). The relevant limits are added to the corresponding materials, such as (*Pyrochlore* or *Pyroch** or $A_2B_2O_7$) for Pyrochlore oxide, (*Perovskite* or *double perovskite* or **perovskite** or $A_2B_2O_6$ or ABO_3 or $A_2B_2O_{5.5}$) for Perovskite oxide, (*Ruddlesden-Popper* or **RP** or RP_{214} or A_2BO_4 or AB_2O_4) for Ruddlesden-Popper oxide. The bibliographic scan was combined with a fuzzy string marked by the symbol "*," which gives a broader set of terms similar to the keywords.

On Dec 2020, English documents published between the years 1990 and 2020 were gathered, with the search focusing on the inclusion of common keywords in the documents. A crucial screening of the retrieved bank was conducted to ensure the accuracy of the data gathered. The selected papers were chosen from the WoS “marked list” and exported in the format “plain text” before being incorporated into ScientoPy (v.2.0.3.7z) to be analysed based on their particular characteristics. The following scientometric parameters were used in this analysis:

Table 1
Various sets of keywords were used to retrieve the published documents in WoS on oxide-based catalysts for water splitting.

Set	Keywords	Results (Number of documents)
#1	TS=(oxide or oxide* or Ruddlesden-Popper or RP or RP_{214} or A_2BO_4 or AB_2O_4 or Perovskite or double-perovskite or *perovskite* or $A_2B_2O_6$ or ABO_3 or $A_2B_2O_{5.5}$ or Pyrochlore or Pyroch* or $A_2B_2O_7$)	1,504,985
#2	TS=(Pyrochlore or Pyroch* or $A_2B_2O_7$)	10,117
#3	TS=(Perovskite or double-perovskite or *perovskite* or $A_2B_2O_6$ or ABO_3 or $A_2B_2O_{5.5}$)	101,085
#4	TS=(Ruddlesden-Popper or RP or RP_{214} or A_2BO_4 or AB_2O_4)	2084
#5	TS=(OER or HER or water splitting or oxygen evolution or hydrogen evolution)	388,685
#6	#1 AND #5	29,761
#7	#2 AND #5	231
#8	#3 AND #5	2944
#9	#4 AND #5	342

(1) category, (2) time, (3) contributing countries, (4) author, (5) published journals, (6) groups, and (5) keywords.

Granted patents were also analysed. The search engine “patents. Google” was chosen as database, and the keywords “*water-splitting*” AND “*photochemical*” or “*electrochemical*” were set. The patents were chosen in “Grant” status. In order to evaluate the possible correlations between articles and patents, a statistical analysis was carried out by using the Pearson correlation coefficient. Further investigations were carried out by evaluating the patent registration centre.

3. Result and discussion

By performing an automatic search in the Web of Science website using a set of keywords, a total of 29761 English documents were collected for the period from Jan 1990 to Dec 2020. through the web of science. The findings obtained by carrying out the research design are described following the scientometric parameters stated in the Methodology section. During the discussion part, pyrochlore oxides, perovskites, and Ruddlesden-Popper oxides are abbreviated as PYs, PEs, and RPs.

3.1. *Overall publication analysis of the oxide-based catalysts*

The publications of oxide-based catalysts are analysed from different aspects, such as document types, country contributions, keywords, and published journals (Fig. 1). It could be determined that the blue line represents of the number of the published documents on oxide-based catalysts for water splitting has grown dramatically since 2012, while the number of the published documents with different types almost remains constant (Fig. 1a and Table S1). The blue line (document type of article) represents the number of the published articles over time is much higher than the number of publications with other types, especially since 2012 (Fig. S1a). In particular, the blue point (reflect document type of article) on the right side of Fig. 1a shows the average number of the published articles per year (2019 and 2020) is more than 4500. This point is also much higher than other points and has accounted for about 30% (in the own corresponding type) of the cumulative number of the published articles since 1990, which indicates the rapid development of research on oxide-based catalysts for water splitting in recent two years. Along with the development of characterisation techniques such as ex-situ/in-situ transmission electron microscope (TEM) and scanning electron microscope (SEM) for the investigation of morphologies (Han et al., 2016), energy-dispersive X-ray spectroscopy (EDX) (Gao et al., 2019), inductively coupled plasma-atomic emission spectroscopy (ICP-AES), X-ray photoelectron spectroscopy (XPS) for the investigation of element existence and oxidation state of materials (Liu et al., 2019), Raman Spectrometers, high-resolution transmission electron microscopy (HRTEM), selected area electron-diffraction (SAED) (Han et al., 2019), and X-ray diffraction (XRD) for the investigation of the phase structure of synthesised catalysts, the research on oxide-based catalysts for water splitting in the recent two years developed quickly. From Figs. 1b and S1b and Table S2, the number of published documents on oxide-based catalysts for water splitting has grown rapidly in China since 2012, while it is only a small increase in other countries. And the number of the published documents in China (blue line) is far greater than that in other countries since 2014, and the average number (2019–2020) accounts for about 50% of the cumulative number of the published articles since 1990. The order (Top

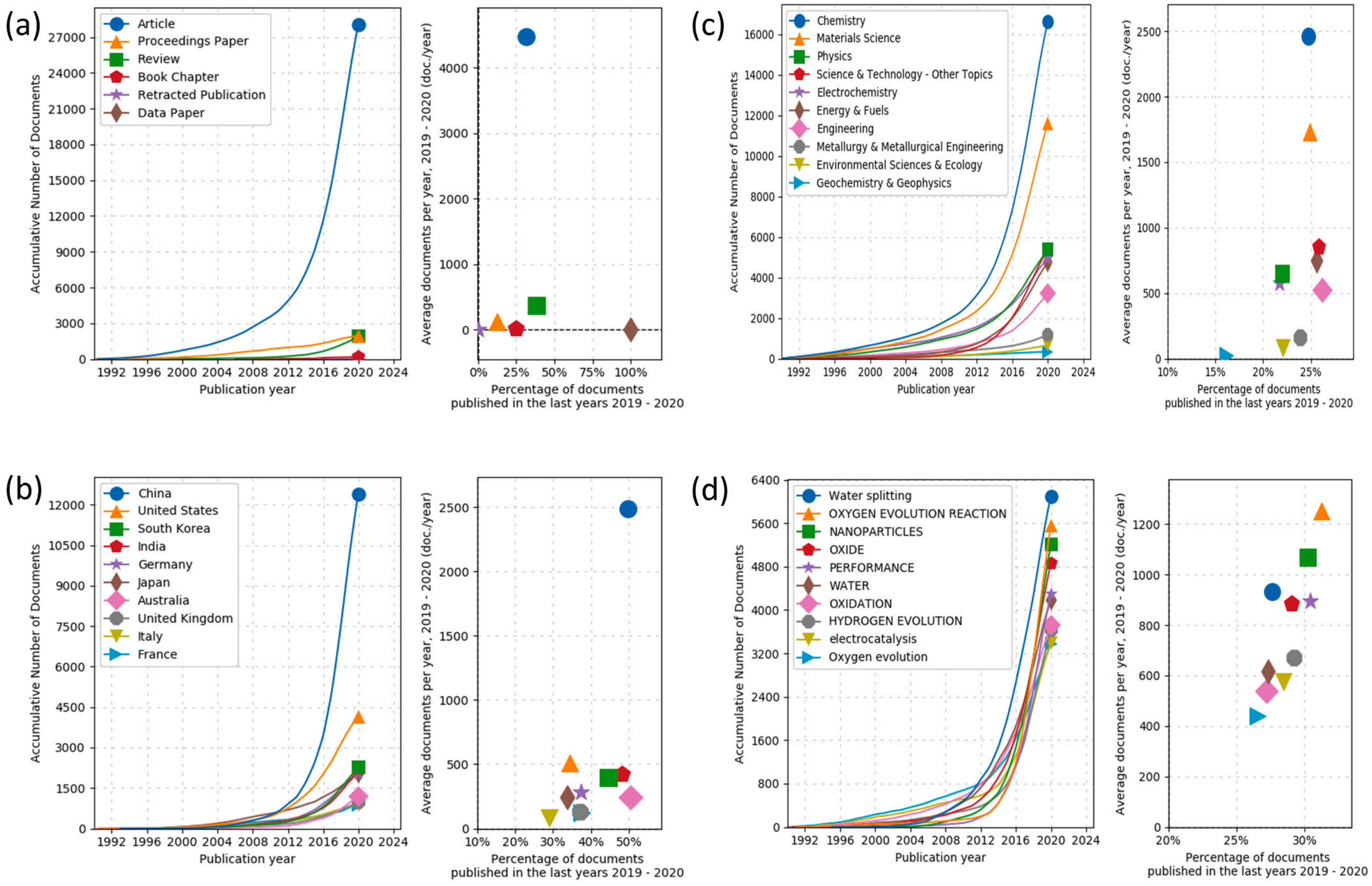

Fig. 1. The sigmoidal pattern with curve fitting of the cumulative number of publications over 1990–2020 on oxide-based catalysts for water splitting from (a) different document types, (b) different countries, (c) categories, and (d) both keywords from the Authors and cited documents. The average documents per year (2019–2020) was listed, with the percentage of the corresponding parent cumulative number were also listed.

10) base on country contributions is roughly listed as: China > United States > South Korea > India > Germany > Japan > Australia > United Kingdom > Italy > France. From Fig. 1c, the analysis on categories, it could be determined that the publications on this topic are connected with Chemistry, Materials Science, and Physics. Recently, more and more attention is paid to the investigations on oxide catalysts for water splitting from materials synthesis to reaction mechanism exploration. For example, Chen et al. reported that during the electrochemical cycling, a surface reconstruction, with Sr and Co leaching, over $SrCo_{0.9}Ir_{0.1}O_{3-\delta}$ occurs (Chen et al., 2019). Such reconstructed surface region, likely contains a high number of structural domains with corner-shared and under-coordinated IrO_x octahedrons, is responsible for the observed high activity; Huang et al. summarised the novel paradigm for catalyst design by overcoming or circumventing the adsorption-energy scaling relations (Z.-F. Huang et al., 2019). The keywords of indexed documents were also analysed in Figs. 1d, S1, S2. The different lines represent the number of the published documents with different keywords over time have a common feature (rapidly developed since 2014). The top-five keywords are “water splitting”, “oxygen evolution reaction”, “electrocatalysis”, “photocatalysis”, and “hydrogen evolution reaction” (from the author, Figs. S1d and S2a and Table S3); “oxide”, “nanoparticles”, “performance”, “water”, and “oxidation” (from the cited documents, Figs. S1e and S2b and Table S4); “water splitting”, “oxygen evolution reaction”, “nanoparticles”, “oxide”, and “performance” (from both author and the cited works, Figs. 1d and S1f and Table S5). These results are associated with the research trend on this topic, which indicates solar energy is potential clean energy to support the water-splitting process, and nanotechnologies are also widely applied for the design of the catalyst in this research direction. The number of documents that contain the keyword “water splitting” or “oxygen evolution reaction” from the author as well as both from the author and cited documents is always relatively larger than other keywords since 2017. The average numbers (2019–2020) of the published documents with different index keywords all occupy more than 32% of the corresponding parent cumulative number since 1990. Figs. S1c and S2c and Table 2 represents the numbers of the published documents on oxide-based catalysts for water splitting in different journals over similar years. Since 2014, they have all grown at a high rate. The average number (2019–2020) takes up more than 18% of the cumulative number of the published documents since 1990 for all journals, agreeing well with the scientific trend of rapid development in 2019–2020 in Fig. 1a. The top-three published peer-review journals are the International Journal of Hydrogen Energy, Journal of Materials Chemistry A, and Electrochimica Acta.

3.2. Distribution of publications from perovskites, Ruddlesden–Popper perovskites, pyrochlore-type oxides

As shown in Fig. 2, the crystal structure of PEs (Fig. 2a), RPs (Fig. 2b) and PYs (Fig. 2c) is becoming more and more complex. The Ruddlesden-Popper oxides are also widely named as layer perovskite oxides because of the correlation on the crystal (Ruddlesden-Popper (A_2BO_4) = Perovskite (ABO_3) +AO layer). The pyrochlore oxides with the general formula of $A_2B_2O_7$ are similar to those of the double perovskites ($A_2B_2O_6$). The numbers of the published article over the years with PEs (Figs. 2d and S3a), RPs (Figs. 2e and S3b) and PYs (Figs. 2f and S3c), are all in a substantial increase since 2012 while the numbers of the published review, proceeding paper, book chapter as well as data paper stay almost the same level. The number of the published article with PEs, RPs and PYs far outweigh the numbers of the published review, proceeding papers, book chapters and data papers since 2012. It is consistent with the overall trend analysis of the number of the published documents on oxide-based catalysts for water splitting in Fig. 1a. The number of published articles from PE in 2019 is more than 420, which is eight times the number in 2012. The number of publications on RPs and PYs in 2019 is also around 8–10 times those in 2012, but one order of magnitude less compared to PEs. From Fig. 2d, about 1000 articles from PEs are published in the last two years (2019–2020), accounting for about 31% of the cumulative number (around 2700) of the published articles since 1990 (Figs. S3a and S3d). The number of published articles on RPs (Fig. 2e) is much less than PEs. It is obviously less than one-tenth of the number of the published article with PEs every year. The number of the published article in the last two years (only around 70) in RP structure takes up 23% of the cumulative number (round 315) of the published articles since 1990 (Figs. S3b and S3e). The blue line in Fig. 2f represents the number of the published articles about PYs; it is slightly choppy, with an overall upward trend, similar to the blue line of the number of documents for RP materials. The number of the published article in PY structure is even smaller than RP one. The number of the published articles in the last two years (2019–2020) with PY structure is about 40, responsible for 16% of the cumulative number of published articles since 1990 (Figs. S3c and S3f).

Table 2
The number of the published documents on oxide-based catalysts for water splitting contributed to different journals.

Source Title	Total number
International Journal of Hydrogen Energy	1,343
Journal of Materials Chemistry A	1,024
Electrochimica Acta	1,012
ACS Applied Materials and Interfaces	731
Journal of Physical Chemistry C	676
Journal of the Electrochemical Society	521
Applied Catalysis B-Environmental	498
Applied Surface Science	486
RSC Advances	455
ACS Catalysis	410

3.3. Contributing countries analysis for perovskites, Ruddlesden–Popper perovskites, and pyrochlore-type oxides

Earth-abundant electrocatalysts can significantly reduce costs while preserving operation and longevity (Mohammed-Ibrahim and Sun, 2019). Rapidly formed metal oxides as electrocatalysts for HER and OER (Kuznetsov et al., 2020) that operate in a broad range of pH mediums (acid, alkaline, neutral, and seawater) have received a lot of attention because they can increase overall performance (Li et al., 2019). The numbers of the published articles over the years with three type structures (PE, RP, PY) grow explosive since 2013, especially in China and United States (Figs. 3 and S4). For other countries listed, the number of published articles with PE structure has increased over the years while is obviously slower than the number in China. The numbers of the published article with PE in China and the United States are greater than the other countries, such as Japan, South Korea, Germany etc. From the cumulative number of publications over 1990–2020 for PEs (Figs. 3a and S4a), RPs (Figs. 3b and S4b) and PYs (Figs. 3c and S4c) with different Countries, it could be determined that the top three countries are China > United States > Japan. Quantitatively speaking, scientists in China contributed more than others in PEs, RPs and PYs-based catalysts for water splitting, agreeing well with the overall trend analysis on oxide catalysts in Fig. 1b. For PEs, until Nov. 2020, the cumulative number of publications for China is about 1050, which is two times that of the USA. The number of publications in Japan is generally half of the that of United States. The percentage of documents published in the last two years (2019–2020) in China is 32%, and it accounts for 25% in the United States.

The Top three Authors who contributed to scientific publications on PEs (Fig. 4a) is Shao, Z.P.>Zhou, W.>Domen, K. The Top two Authors of Shao, Z.P. and Zhou, W. coming from the same group in Nanjing Tech University have published more than 75 articles since 1990, of which greater than 42% were published in 2019–2020. The number of the published documents with RP structure also takes one-tenth less than

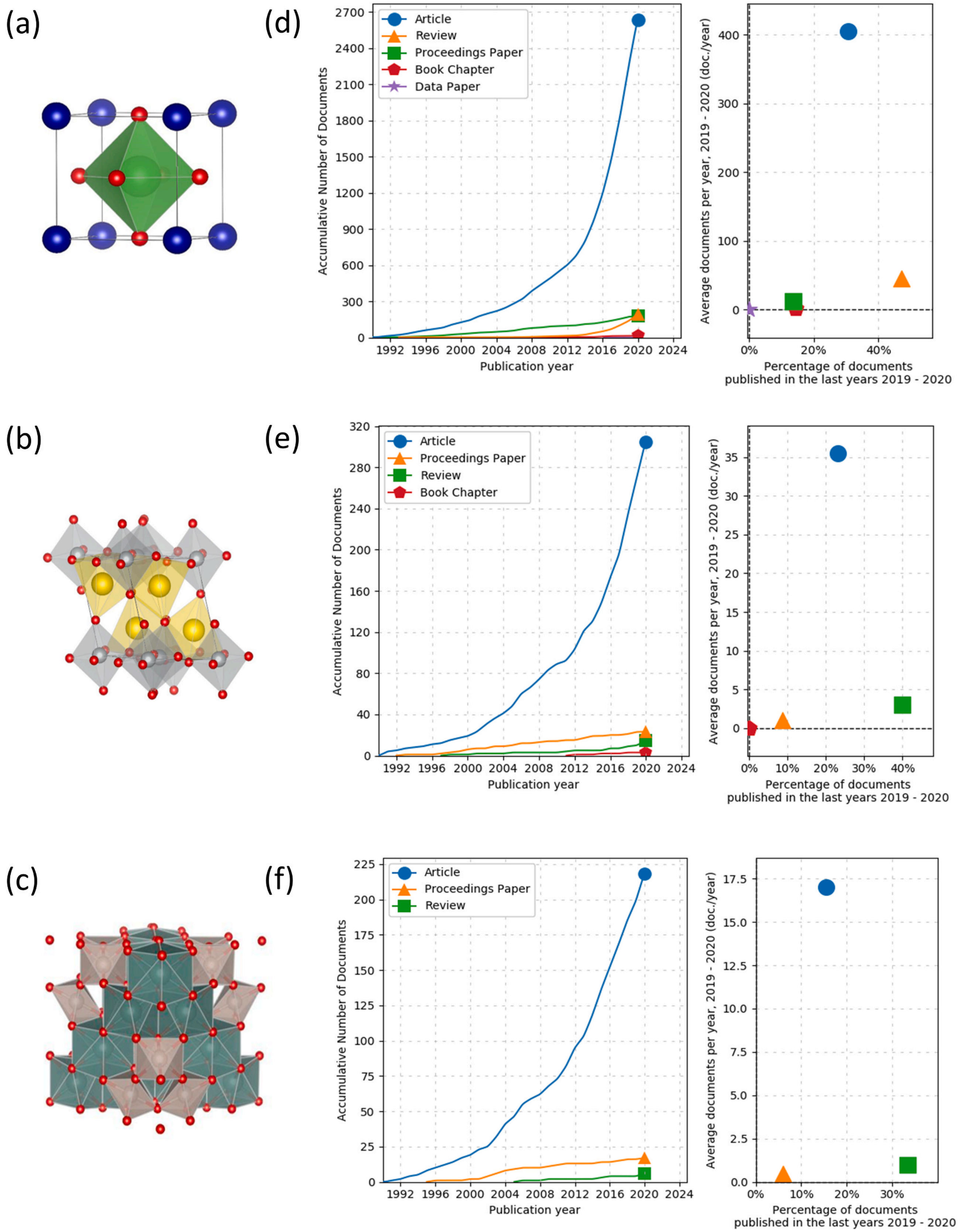

Fig. 2. The crystal structure of (a) perovskites (PEs, ABO_3), (b) Ruddlesden-Popper oxides (RPs, A_2BO_4), (c) Pyrochlore-type oxides (PYs, $A_2B_2O_7$). The sigmoidal pattern with curve fitting of the cumulative number of publications over 1990–2020 on with different document types from PEs (d), RP oxides (e), PY oxides (f). The average documents per year (2019–2020) was listed, with the percentage of the corresponding parent cumulative number were also listed.

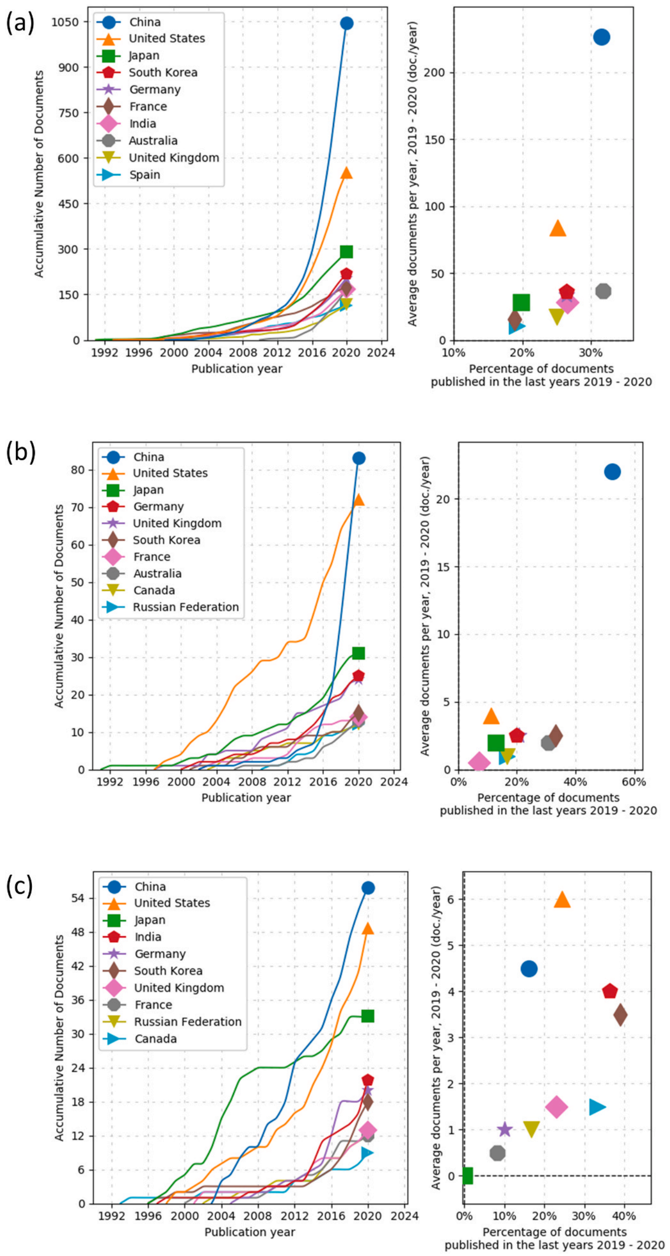

Fig. 3. Contributions from different countries around the world to the development of scientific documentation on water splitting, the sigmoidal pattern with curve fitting of the cumulative number of publications over 1990–2020 for PEs (a), RPs (b), PYs (c).

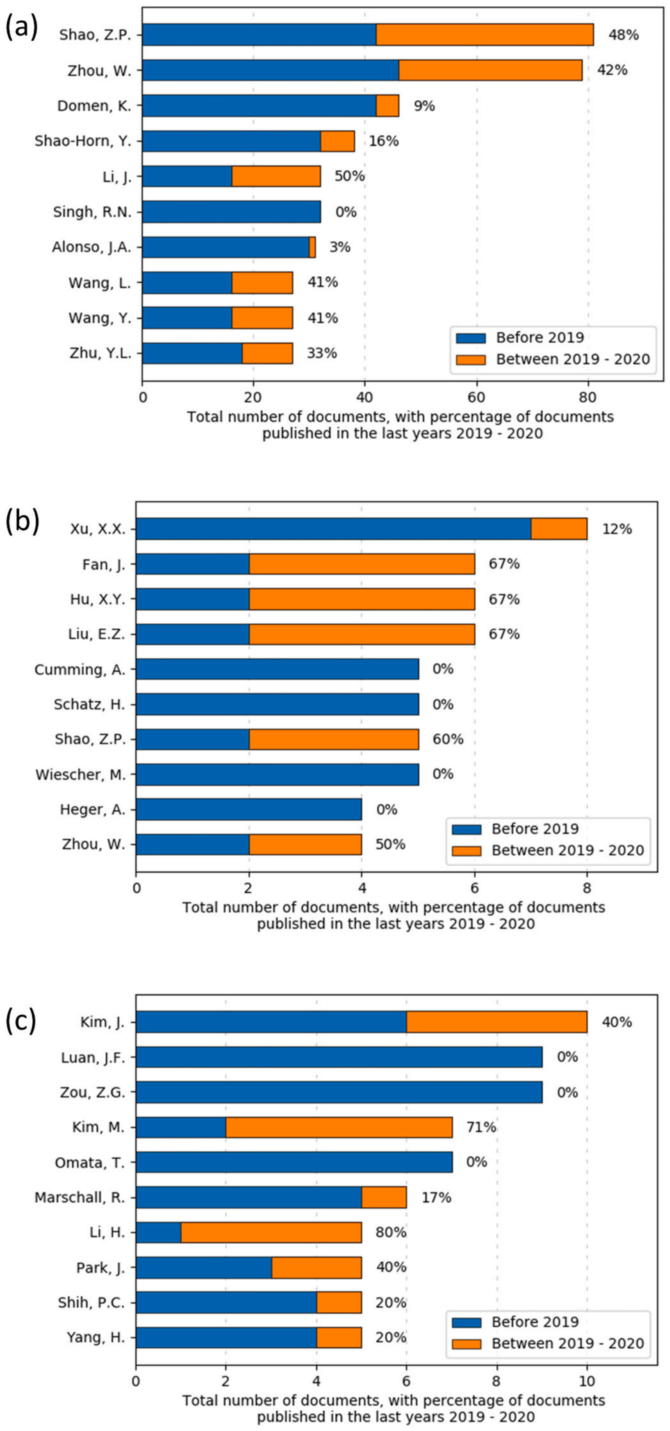

Fig. 4. The Top 10 authors contributed in scientific publications on oxide catalysts water splitting based on PEs (a), RPs (b) and PYs (c).

the number of PEs over years, which coincides with Fig. 3a and b. The number of published articles with RP structure in China has increased rapidly since 2014 and is up in a wave in the United States. The number of the published article over the years in other countries with RPs stays the same level. The difference with the cumulative number of publications over 1990–2020 for RPs is far less than the difference with PEs. About 42% of the cumulative number of publications over 1990–2020 in China is published in 2019–2020, and the number takes up about 12% in the USA (Fig. 3b). For the USA and Japan, it is less than 18%. The Top three Authors who contributed to scientific publications on RPs (Fig. 4b) is Xu, X.X.> Fan, J.> Hu, X.Y. The top one author of Xu, X. published eight articles since 1990 (actually since 2015), and the number of documents published in 2019–2020 only takes up 12%. And the number of documents published in 2019–2020 for Fan, J. and Hu, X.Y accounts for 67% of the corresponding cumulative number. Different curves in Fig. S4c, standing for the numbers of the published documents over the years with PYs in different countries, all have a large amplitude of vibration, and the difference between the curves is small. This could be attributed to the small number of publications on PY structure. The cumulative number of publications over 1990–2020 for PYs in China is about 55, one-twentieth of PEs (1050 in Fig. 3a) and three-fifths of RPs (84 in Fig. 3b). The percentage of documents published about PYs for water splitting in 2019–2020 in China is about 18% which is small than 25% in the United States. The top three authors contributed to scientific publications on PYs (Fig. 4c) Kim, J.>Luan, J.F.>Zou, Z.G. The top author Kim J. who has published 10 articles since 1990 and the number of documents published in 2019–2020 takes up 40%, while the number of documents published in 2019–2020 for Luan, J.F and Zou, Z.G. is zero.

3.4. Journal contributions and categories analysis for perovskites, Ruddlesden–Popper perovskites and pyrochlore-type oxides

The top three journals base on the cumulative number of publications on water splitting from the overall oxides (Fig. 5a) are in descending order International Journal of Hydrogen Energy, Journal of Materials Chemistry and Electrochimica Acta. For PE-based catalysts (Fig. 5b), the publications are consistent with the overall oxide catalysts with the same top three journal contributions. The percentages with different journals from PEs in the last two years (2019–2020) are all more than 20%. Meanwhile, for RPs (Fig. 5c) and PYs (Fig. 5d), the top three hottest Journals are Astrophysical Journal, Applied Catalysis B: Environment, International Journal of Hydrogen Energy and Journal of Solid-State Chemistry, International Journal of Hydrogen Energy, Applied Catalysis B: Environment. The total number of documents of Astrophysical Journal from RPs is only ten which is one-fourteenth of the number of International Journal of Hydrogen Energy from PEs (close to 140). It also indicates that hydrogen production is a cut-edging research direction, as the International Journal of Hydrogen Energy is always the popular journal for these catalysts. The investigations on clean energy production are associated with materials science from material synthesis to material analyses (e g. Journal of Materials Chemistry), environment science from hydrogen production to environmental application (e g. Applied Catalysis B: Environment), electrochemistry from electrochemical water splitting to photoelectrochemical water splitting (e g. Electrochimica Acta). The development of this technique to produce hydrogen is also a new multidiscipline crossing edge direction.

The categories of the publications are also summarised. The different figures with different curves represent the numbers of documents with different categories over time from the overall oxides, PEs, RPs and PYs are similar (Fig. S5). The number of publications on chemistry from oxides, PEs, RPs and PYs, is the largest and the growth rate also is the fastest since 2012. The number of publications over the years from different aspects of materials science, electrochemistry, science and technology (other topics), energy and fuels, physics, and engineering has also increased significantly, while in metallurgy and metallurgical engineering, environmental science and technology stay the same. The top one of the percentages of documents published in the last two years (2019–2020) on different categories from oxides, PEs, RPs and PYs are the same in material science. While the top three categories are the same of oxides, PEs, RPs and PYs, and also display in the same order of chemistry > material science > physics. It indicates that a lot of works are focused on materials synthesis (connected with materials science and physics) and also performance optimisation (connected with chemistry). The percentages of documents published in 2019–2020 with different journals from the overall oxides are all more than 17% (Fig. 6a). The percentages for the top three are even more than 29%. The cumulative number of documents from the overall oxides with chemistry and materials science is more than 11,000, far greater than other categories. The percentages of documents published in the last years 2019–2020 from the overall oxides with chemistry and materials science are 29% and 30%. In different categories, most percentages of documents indexed in the last two years (2019–2020) for the overall oxides are between 23% and 30%. From Fig. 6b, it could be determined that the percentages with different journals from PEs are all more than 18%, and the top five categories are more than 30%. Most percentages for PEs in different categories range from 30% to 43%. The difference in the number of documents with categories from RP oxide is smaller than the difference from PEs. Most percentages for RPs in different categories are range from 32% to 60% (Fig. 6c). The curves in Fig. 6d represent the numbers of documents with different categories over time from PYs are entirely different from the other. They have a large amplitude of oscillation. Meanwhile, percentages (last two years) for PYs in different categories are almost in a range from 7% to 17% (Fig. 6a).

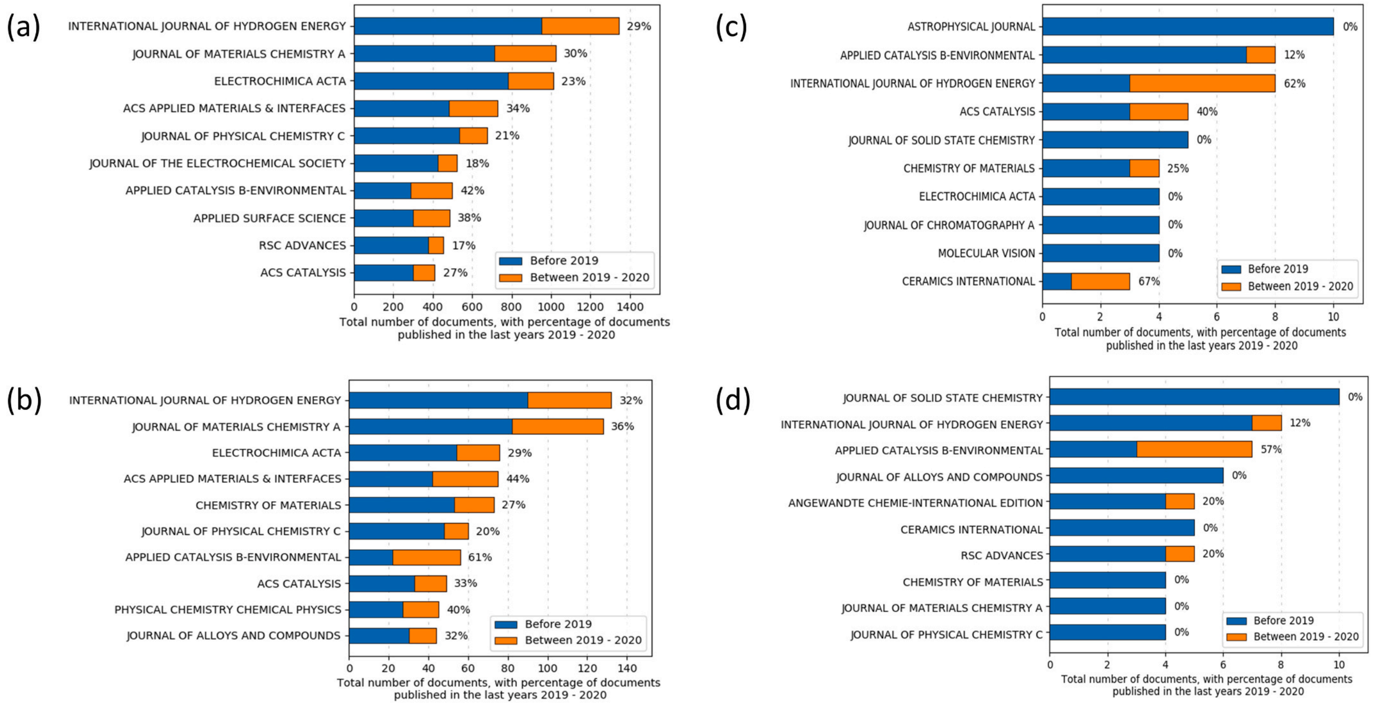

Fig. 5. Analysis based on the cumulative number and even with the percentage of documents indexed in the last two years (2019–2020) on water splitting from the overall oxides (a), PEs (b), RPs (c), PYs (d).

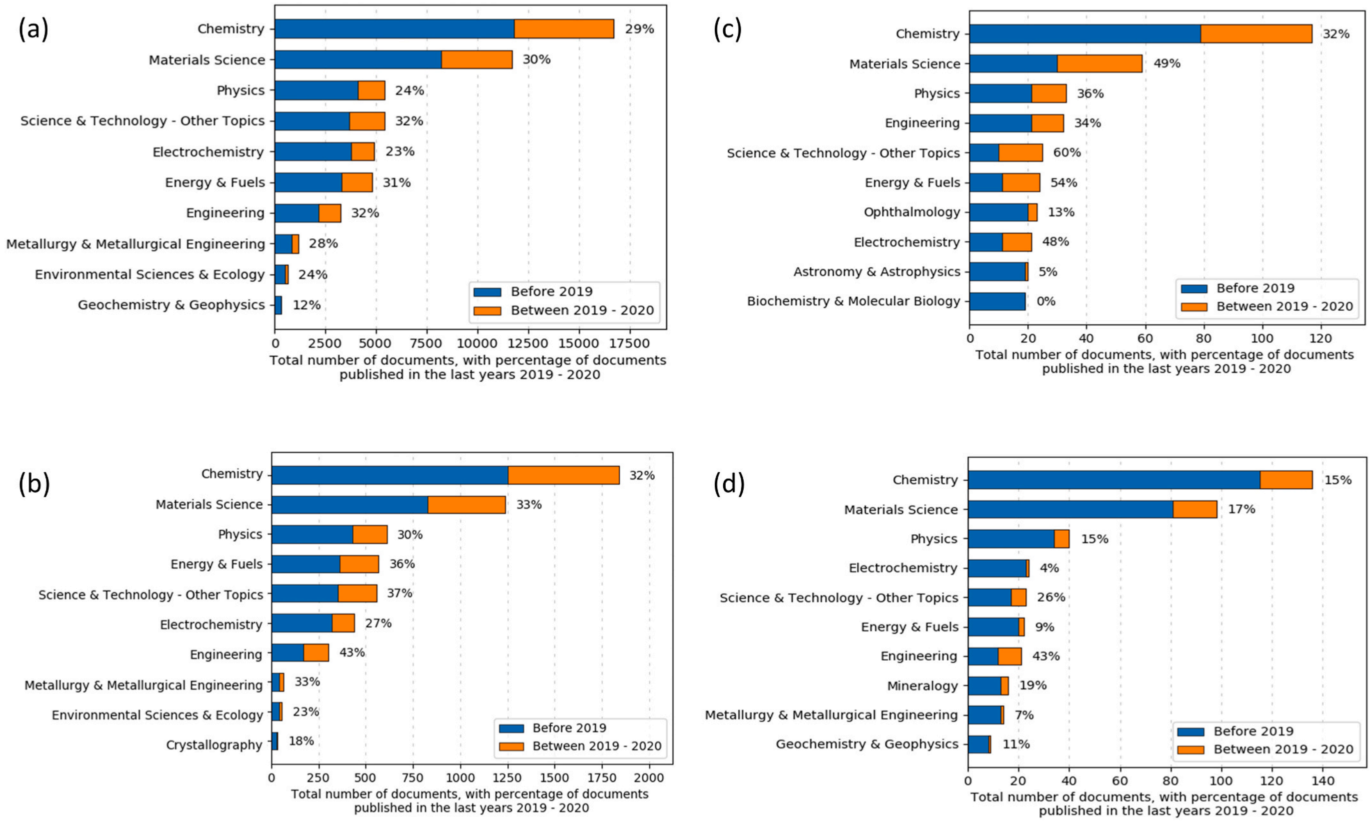

Fig. 6. Categories analysis of the published documents, the cumulative number of documents with different categories and even with the percentage of documents indexed in the last two years (2019–2020) for the overall oxides (a), PEs (b), RPs (c), PYs (d).

3.5. *Keywords analysis for perovskites, Ruddlesden–Popper perovskites and pyrochlore-type oxides*

The top-three keywords from PEs are "perovskite", "oxygen evolution reaction" and "water splitting" (from the Authors, Fig. 7a); "evolution", "water" and "performance" (from the cited documents, Fig. 8a); "perovskite", "evolution" and "water" (from both Authors and the cited papers, Fig. S6a). The total numbers of documents contain different keywords for PEs from the Authors range from 70 to 280, with the percentage of documents published in 2019–2020 changes from 14% to 51%. The range of the total numbers for PEs from the cited documents is from 6 to 13, with the range of percentage is 0–62%. The top keywords from PE structure (from the cited documents), "retinitis pigmentosa", has a percentage of 14%, while percentages for "photocatalysis "and "red phosphorus" are 54% and 62%. The total numbers for PE structure from both Authors and the cited works range from 5 to 23, together with the percentage of documents published in 2019–2020 change from 0 to 30%. Photocatalysis is one of the most important keywords in perovskite oxide catalysts, which reflects the importance of the utilisation of solar energy. For example, Kato's team investigated the role of Ag^+ in the band structures and photocatalytic properties of $AgMO_3$ (M: Ta and Nb) with the perovskite structure (Kato et al., 2002). Tan's group reported that adjust the oxygen vacancy could enhance the photocatalytic activity of perovskite $SrTiO_3$ (Tan et al., 2014); Wang et al. reported a novel single-junction cathodic approach for stable unassisted solar water splitting (Wang et al., 2019). van de Krol's team summarised a faster path to solar water splitting (van de Krol, 2020). Based on a special chemical-physical property of phosphides, red phosphorus is usually a phosphorus source for the further modification of oxide catalysts to prepare transition metal phosphides as heterogeneous electrocatalysts for water splitting (Wang et al., 2017), red phosphorus is another important keyword.

Five of ten keywords from both Authors and the cited works for PEs have a percentage of zero. The top-two keywords for PEs from the Authors (Fig. 7a), from the cited documents (Fig. 7b), from both authors and the cited works (Fig. S6a), are the same, which are "evolution" and "water". The total number of documents from the authors ranges between 220 and 420 with various keywords, with percentages ranging from 24 to 47%. The range of the total numbers for RPs from the cited documents changes from 11 to 30, and the percentage covers from 0 to 69% (Fig. 8a). Eight of ten keywords from the cited documents for RPs have percentages that are greater than 41% (Fig. 8b). The total numbers for RPs from both author and the cited works range from 18 to 42 together with the percentage varies from 0 to 40%. Seven of ten keywords from both Authors and the cited works for RPs have percentages which are small than 20% (Fig. S6b). The top-three keywords from PY oxide are "pyrochlore", "evolution" and "water" (from the Authors, Fig. 7c); "evolution", "water" and "red phosphorus" (from the cited documents, Fig. 8c); "evolution", "pyrochlore" and "water" (from both Authors and the cited works, Fig. S6c). The total numbers with different keywords for PY structure from the Authors change from 220 to 610, with the percentage of documents varies from 30% to 50%. The top keyword from the author, "pyrochlore", has a total number of more than 600, while the total number of remaining keywords is less than 420. The total numbers for PYs from the cited documents range from 14 to 30, with percentages from 14% to 65%. Seven of ten keywords' percentages from the cited works for PYs are more than 40%. The total numbers for PY structure from both Authors and the cited works are from 18 to 44, and the percentage changes from 5% to 39%. Seven out of ten keywords' percentages from both Authors and the cited works are less than 20%.

Noteworthy that "photocatalysis" is the hot keyword in all these three types oxide catalysts (PEs, RPs, PYs), which indicates the importance of the utilisation of solar energy. Making good use of abundant, inexhaustible solar energy is another way to achieve sustainable

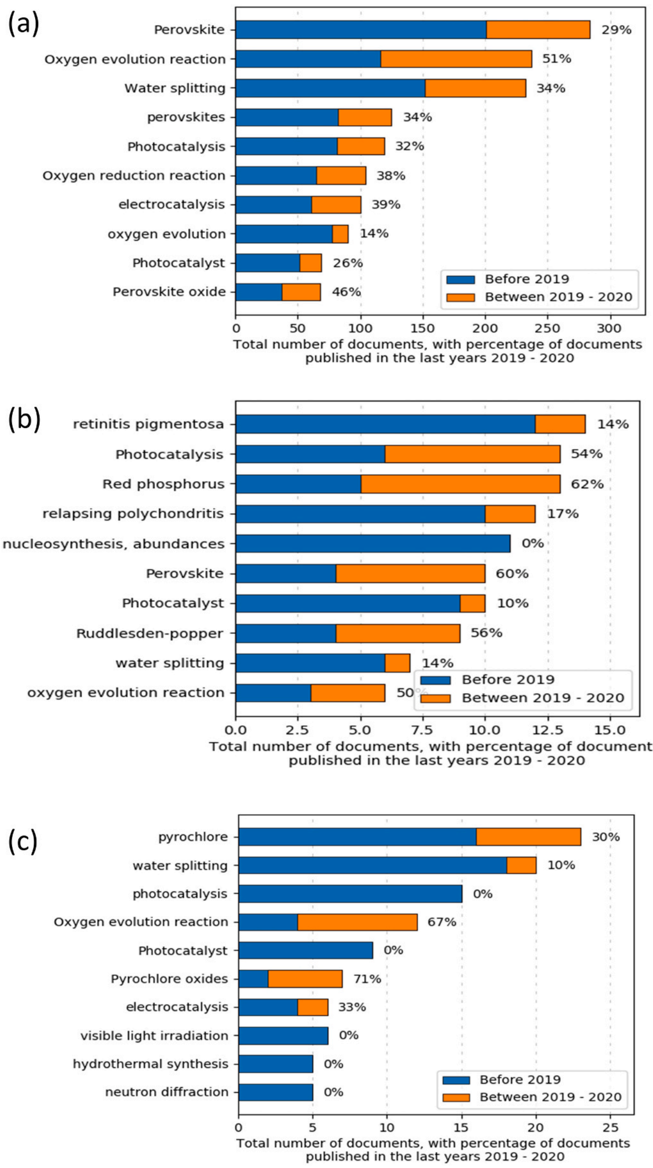

Fig. 7. Keywords analysis from the Authors for PEs (a), RPs (b) and PYs (c) based catalysts for water splitting; The percentages of the keywords used in the last two years (2019–2020) to the parent cumulative number were also listed.

hydrogen output. The photo (electro)chemical water splitting for the conversion of solar energy to chemical energy (hydrogen) is one of the most exciting paths for long-term energy supply. Since Fujishima's work on TiO_2-driven photoelectrochemical hydrogen production (Fujishima and Honda, 1972), lots of semiconductor-based photocatalysts have been developed, especially those that operate under visible light irradiation to take advantage of a large portion of sunlight, such as $NaTaO_3$ (Kudo and Kato, 2000), $SrTiO_3$ (Wang et al., 2015), $BaTiO_3$ (Zhang, G. et al., 2016). The most popular photocatalysts are native and mixed metal oxides (Liu et al., 2021), (oxy)nitrides (Concina et al., 2017), sulphides (Benck et al., 2014), metal-carbon compounds (Liu et al., 2015), tricomponent metal oxides (Zhang et al., 2019), tantalum-based semiconductors (Zhang et al., 2014).

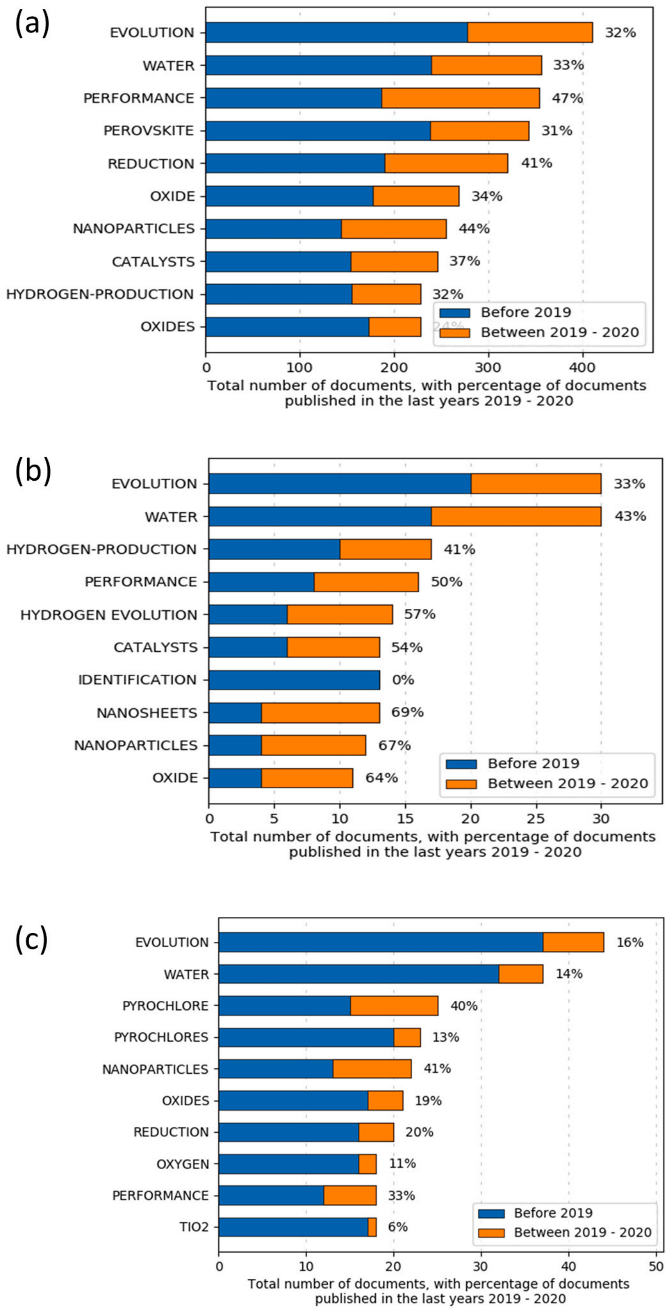

Fig. 8. The cited keywords for PEs (a), RPs (b) and PYs (c) based catalysts for water splitting. The percentages of the keywords used in the last two years (2019–2020) to the parent cumulative number were also listed.

3.6. Scientific trends

The findings of this report show that the majority of the scientific activities that culminated in indexed publications occurred mostly after the year 2000. Only a few publications were published between the years 2000 and 2010, showing that research expertise in this field was only getting started. Several significant research results have led to the rise of oxide electrocatalysts (Suntivich et al., 2011a) and also (Suntivich et al., 2011b). The "onset year" of oxide-based catalysts for water splitting is 2010 (Fig. 1a), while that of PE catalysts appears in 2012 (Fig. 2d). Noteworthy the "onset year" of RPs and PYs are indefinable (close to 2016 for both of them), which could be attributed to the that the number of publications on RPs and PYs is in one order of magnitude less compared to PEs (Fig. 2). It indicates that the latter two oxide systems (RPs and PYs) are relatively new research systems to investigate and attract more and more attention.

In ABO_3 of PE structure, A-site is usually occupied by alkali metals such as La, Sr, Ba, Ca, while B-site are filled with transition metals such as Co, Fe, Mn, Ni. Up to now, perovskite oxides have been widely investigated for electrochemical water splitting (Suntivich et al., 2011b) and photoelectrochemical water splitting (Markus et al., 2017) in alkaline environments. However, little work reported on the use of perovskite oxide for water splitting in acidic media because of the weak resistant of these metals to acids. A similar phenomenon is also observed for the layer perovskites and RPs oxides. The several groups tried to dope noble metals (Ru, Ir, Pt) into a crystal of perovskite and Ruddlesden-Popper perovskites to develop acidic resistance catalysts for water splitting in acidic media. This phenomenon does not exist for some catalysts with PY structure ($A_2B_2O_7$), as the A-site would be occupied by blunt metal and even nonmetallic elements. For example, $Y_2Ru(Ir)_2O_7$ has been proved with excellent activity and strong stability compared to commercial RuO_2 and IrO_2 for OER in acid media (Kim et al., 2017). From Fig. 2, it could be determined that the investigation on RP and PY systems is just beginning to be studied.

Fig. S7 demonstrate the results of the authors' contribution in terms of number of published documents on PEs, RPs and PYs-based catalysts for water splitting. As evident in the outcomes of Citespace and Python, Shao-hong Yang, Rn Singh, and Yunfang Huang are the leading contributors in field of PEs, Xiaomin Xu, Fan Jun, and Enzhou Liu are the leading contributors for RPs, while Jooheon Kim, Jin Kim, and ZG Zou are the leading contributors for PYs. Regarding the publishing sources based on WoS database, Journal of Materials Chemistry A (ISSN: 2050–7496), Journal of the American Chemical Society (ISSN: 0011–9164), and Chemical Communications (ISSN: 1364-548X) are the most active journals in this field for PE (Fig. 9a); Chemistry of Materials (ISSN: 1520–5002), Journal of the American Chemical Society (ISSN: 0011–9164), and Nature (ISSN: 1476–4687) are the most active journals for RPs (Fig. 9b); Chemistry of Materials (ISSN: 1520–5002), Journal of the American Chemical Society (ISSN: 0011–9164), and Journal of Solid State Chemistry (ISSN: 0022–4596) are the most active journals for PYs (Fig. 9c). Overall, Chemistry of Materials, Journal of the American Chemical Society, Angewandte Chemie International Edition, and Journal of Materials Chemistry A are popular journals for all these three kinds of oxides. According to Fig. S8, "perovskite", "catalyst", and "evolution" are the main keywords of the scientific literature in the field of PEs; "hydrogen production", "photocatalysis" and "evolution" are the main keywords for RPs; "catalyst", "evolution", and "electrocatalyst" are the main keywords for PYs, which demonstrates that the application of oxide-based catalysts for water splitting is currently a hot topic. The RPs-based catalysts were widely used for photocatalysis, which is different from PEs and PYs.

Fig. S9a shows the annual registration of patents, and the trend appears to be similar to the publications; in fact, the two parameters are strongly correlated ($r = 0.945$), as also previously reported in previous studies. S.A. Meo found in Europe a positive correlation ($r = 0.76$) between the publications and the patents registered (Meo and Usmani, 2014). About water splitting, the correlation appears higher and could be due to the sector's fast growth in a rapid increase of publications and patents. The number of patents per country shows an increasing trend over time (US (771) > CN (669) > JP (495) > EP (212) >KR (180)). Fig. S9c clearly highlights that the number of patents filed in CN in recent years has grown exponentially and much faster than in other countries. A positive correlation can be found comparing the number of patents with that of publications from each country, although the strength of such correlation quite differs ($r_{MIN} = 0.82$; $r_{MAX} = 0.95$). The publication/patent ratio ranges from 6.6:1 in the USA to 24.3:1 in Europe. The main reason for the different ratios could be due to the R and D strategies of the countries. The trends of patents based on both photochemical and electrochemical treatment were found to be similar to those of publications. The photochemical patents are delayed a few years compared to electrochemical patents as such process is more recent. Both processes show exponential growths in Fig. S9b. The water splitting by using electro/photochemical methods represents one of the areas for future research and industrial developments. A classification of the patents was carried out according to the associated Cooperative

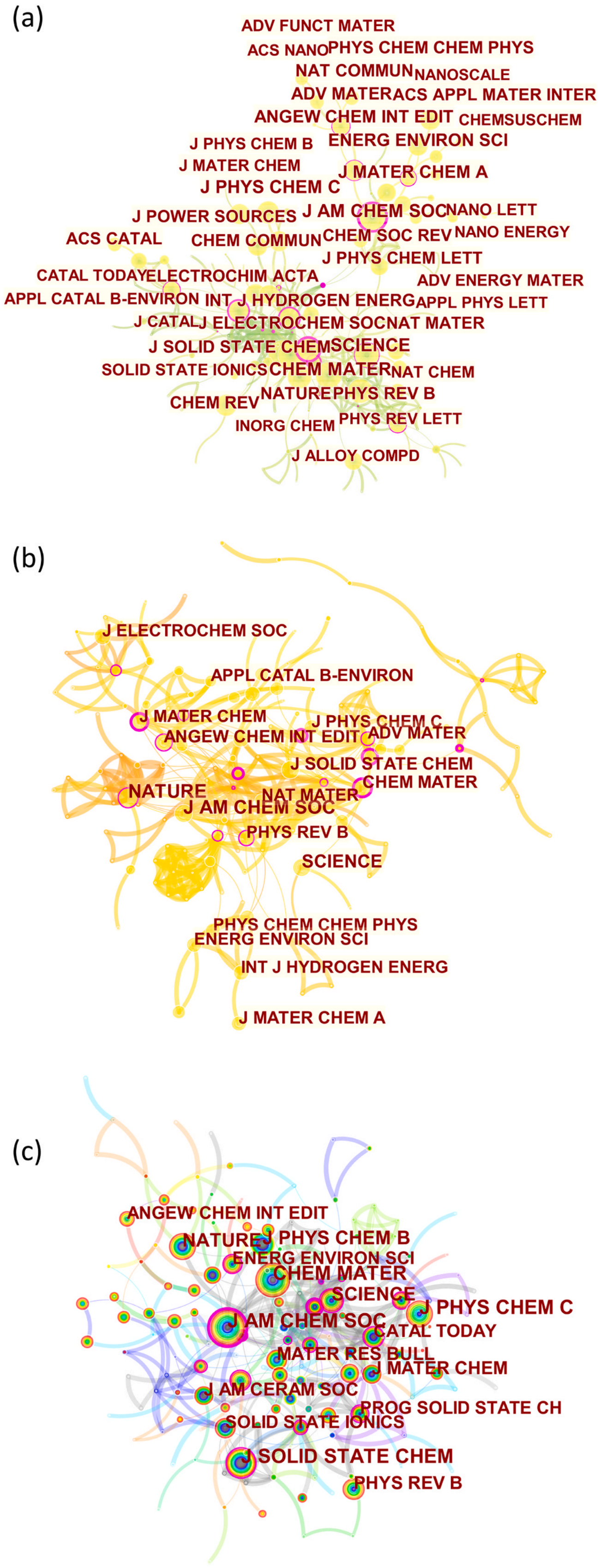

Fig. 9. Contributions of various sources in terms of citations received for the publications on PEs (a), RPs (b) and PYs (c) based catalysts for water splitting. The analysis was performed using CiteSpace.

Patent Classification: more than 50% of the current patent applications have repercussions on the environmental field. They are aimed at reducing greenhouse gases and mitigating climate change. The applications in the fields of inorganic chemistry, water treatment and nanotechnology are present as well. This wide range of applications also in a different area is likely due to the characteristic of water splitting of being capable of predicting the production of hydrogen on-site. The applications have also been found in biomedical, agricultural and solar energy fields, as demonstrated by the patents registered by companies operating in these fields (*i.e.* Nanosys, *Inc*., Groupe Sante Devonian *Inc.*).

The citation explosion, which is a metric for the number of referenced references, is a good measure, will show the most productive field of research within a given time span. The most cited references for the overall oxide-based catalyst (top 10) (Table S6), PE-based catalysts (top 10) (Table S7), RP-based catalysts (top 5) (Table S8), and PY-based catalysts (top 5) (Table S9) were detailly summarised in Supporting Information. To explore the evolutions of the four citation burst groups, Fig. 10 represents the citation histories of the top sources with the largest citation bursts. The references in the first category (overall oxides) have received a significant number of citations since 2001, especially after 2009. This phenomenon reveals oxide-based catalysts is still a research focus. The increased citations were observed for PEs since 2012, which is connected with the several landmark articles published in that year. On perovskite oxides for ORR (Suntivich et al., 2011a), and perovskite oxides for OER (Suntivich et al., 2011b). It could be observed that the citation numbers for articles of RPs and PYs were an order of magnitude lower compared to PEs. Thus, the investigation on RPs and PYs for water splitting is the further direction, which needs more attention.

3.7. Future research directions

Based on the detailed analyses on the publications in recent two years, the published works are mainly focused on materials science, chemistry, and physics, which is connected to catalyst synthesis, and electrochemical/photoelectrochemical water splitting performance. The mechanism investigation on the reaction and material property was also associated with various physical properties (Feng et al., 2014). From a reaction aspect, thermodynamics and kinetics are very important factors. Form catalyst aspect, crystal structure, atomic structure, electron orbit, oxidation state, and oxygen vacancy are the widely investigated factors that are connected to the final water splitting performance. Recently, a lot of composite catalysts were synthesised together with cut-edging materials such as C_3N_4, MXene. Noteworthy that most published works in recent two years in this scientific research area are still focused on material synthesis and normal performance optimisation. The works on mechanism investigation are still taking a very low percentage. To further develop oxide-based catalysts for water splitting, for more study in this area, the following research directions are suggested.

a) The oxide-based catalysts, especially perovskites, have been widely investigated for water splitting in alkaline media. The guidance of mechanism on developing high stable and effective oxides catalysts in acids media has not yet been well organised. The effect of acid conditions on the physical properties of oxide catalysts, which can

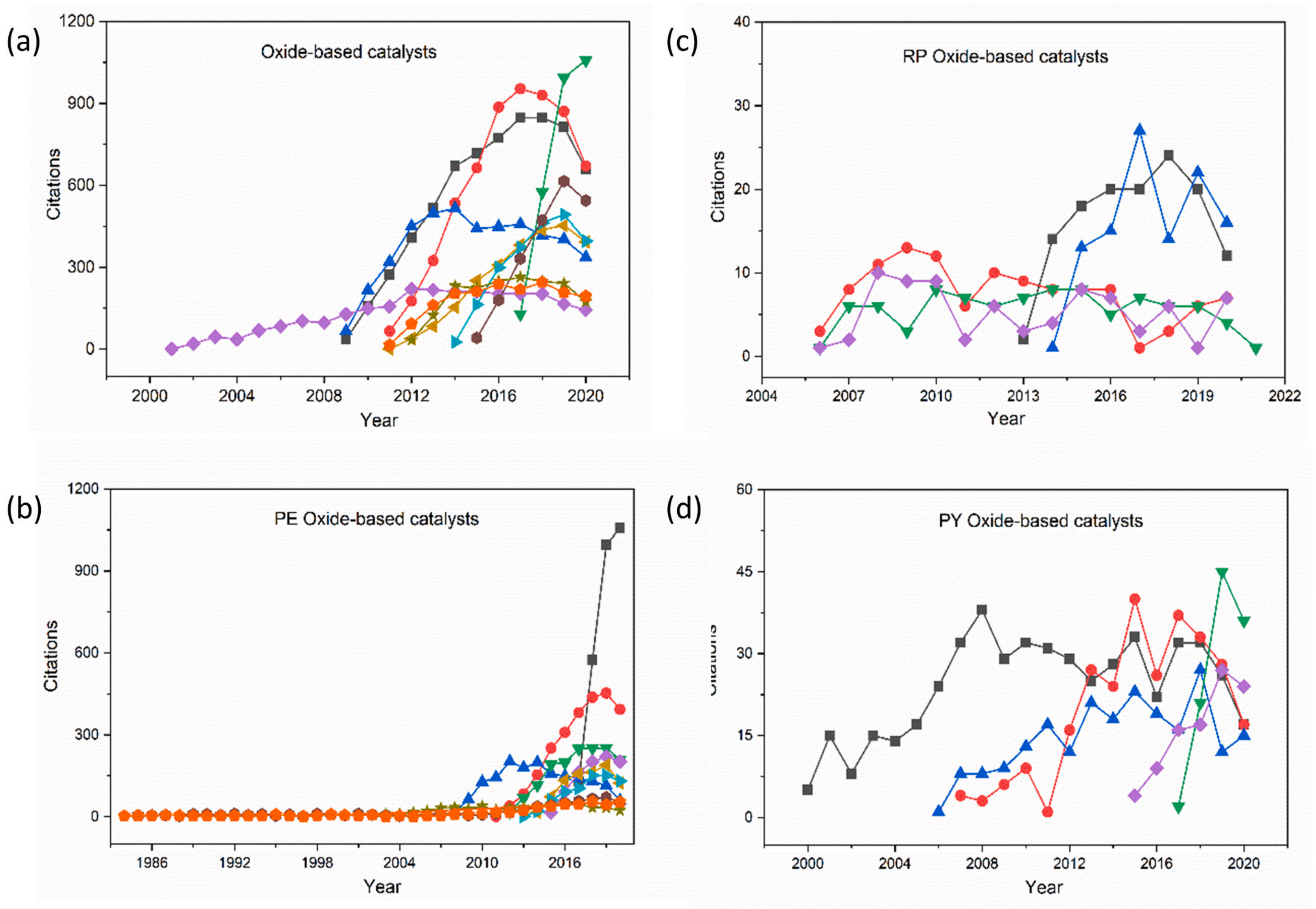

Fig. 10. Citation records for the top 5 or 10 sources for the most significant citation bursts. (a) Overall oxide-based catalysts (top 10 references), (b) PE-based catalysts (top 10 references), (c) RP -based catalysts (top 5 references), (d) PY-based catalysts (top 5 references).

influence the crystal structure and the stability and activity, should be also be investigated.

b) Further research is strongly recommended to optimise the transition-air separating metal oxides considering the variables such as the ionic radii, the electronegativity, the piezoelectric effect, the photoelectric effect etc.
c) Almost no literature of oxide-based catalysts has been reported for water splitting in neutral electrolytes. Thus, more attention should be paid to the investigation of developing suitable oxide catalysts for water splitting in neutral electrolytes.
d) While seawater is the most abundant aqueous electrolyte feedstock on the planet, incorporating it into the water-splitting process poses numerous challenges. The investigation of oxide-based catalysts for seawater splitting is another blank spot.
e) With the improvement of scientific research methods (e.g., the in-situ characterisation analysis), the investigation of the reaction mechanism of oxide-based catalysts for water splitting would be more meaningful.

4. Conclusions

This research was carried out from the various perspectives, and the detailed and multi-perspective overview of the research on oxide-based catalysts for water splitting was given. As water-splitting catalysts, various transition-metal oxide-based binary and ternary perovskites, spinel, ruddlesden–popper, pyrochlore, layered compounds, and other oxides have recently been studied. To boost the catalytic properties of oxide-based electrocatalysts, defect engineering, elemental doping, noble metal coating, morphological nanostructuring, core-shell forming, carbon coating of the catalyst, and organic-inorganic hybrid formation has all been used extensively.

Up to now, a total of 29,761 publications, between Jan 1990 and Dec 2020, have been obtained from the Web of Science website on the research topic of oxide-based catalysts for water splitting. The results of this work show that only a few publications were published between 2000 and 2010, showing that the investigation expertise in this field was only getting started. The number of publications on perovskite oxide catalysts (2,944) is one order of magnitude higher than ruddlesden–popper oxides (342) and pyrochlore oxides catalysts (231), which indicates that the latter two kinds of oxide systems (ruddlesden–popper and pyrochlore) are relatively new research systems to be investigated and attract more and more attention. The “onset year” of oxide-based catalysts for water splitting is 2010, while that of perovskite oxide catalysts appears in 2012. Noteworthy the “onset year” of ruddlesden–popper oxides and pyrochlore oxides are only around 2016, as these kinds of oxide catalysts display potential to overcome the weak resistance to acid medium.

This paper has assisted stakeholders in understanding the global features and dynamics in oxide catalyst research and patents. The electrochemical catalytic and photocatalytic water splitting are one of the future research trends in renewable energy as they are cost-effective, sustainable and environmentally friendly processes. Before a widespread scale application for hydrogen production using water and sunlight, hindered mainly by the low activity and short-term stability of the actual catalysts and the intermittent character of solar energy, possible future water splitting in different scientific and technological fields should be considered. Among the key areas, biomedicine, aircraft and ship engineering and metallurgy are most promising for the entry of sustainable green hydrogen production and use. For example, in biomedical applications, hydrogen and oxygen produced through catalytic water splitting could be used for “on-site” treating different diseases, such as reducing the local inflammation (hydrogen) and overcoming hypoxia (oxygen). The solar “on-site” generation of hydrogen for commercial aircraft/ship engine applications might revolutionise the future in the air and marine transportation business either in terms of aircraft/ship configurations and safety or from the point of view of economic, geopolitical and environmental implications. The catalytic water splitting for “on-site” production of hydrogen in steelmaking processes using ferrous starting materials and wastes of ferroalloy/aluminium industries may considerably reduce carbon dioxide emissions and favour the growth of new cost-effective metallurgical technology. The increase of interest for (photo)-electrocatalytic production of hydrogen from water in these fields might promote the development of cheaper light-driven overall systems (catalysts and devices) for further improving the efficiency of water-splitting processes and realising a large-scale sustainable technology based on hydrogen energy.

CRediT authorship contribution statement

Ning Han: Conceptualization, Investigation, Visualization, Data curation, Writing – original draft. **Marco Race:** Formal analysis, Investigation, Visualization, Data curation, Supervision, Writing – original draft. **Wei Zhang:** Visualization, Writing – review & editing. **Raffaele Marotta:** Visualization, Writing – review & editing. **Chi Zhang:** Visualization, Writing – review & editing. **Awais Bokhari:** Supervision, Writing – review & editing. **Jiří Jaromír Klemeš:** Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2021.128544.

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