

# From Big to Strong: Growth of the Asian Laser-induced Breakdown Spectroscopy Community

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Laser-induced breakdown spectroscopy (LIBS), firstly proposed in 1962 as Brech and Cross<sup>[1]</sup> successfully detected the plasma emission induced by a ruby laser, has attracted more and more attention in both academia and industry due to its unique analytical features such as little or no sample preparation, simultaneous multi-elemental analysis, and remote sensing etc.<sup>[2-4]</sup>. Restrained from the high-cost and poor reliability of instruments back then, the research popularity of LIBS declined quickly after a few years of initial mania of LIBS study. Since the 1990s, benefiting from the significant development of the hardware setups including laser, spectrometer, and ICCD, the “LIBS fever” re-emerged with continuous progress achieved in various applications as well as fundamental studies for the past two decades. In 2004, James D. Winefordner, a prestigious analytical scientist, crowned LIBS as a “future superstar” for chemical analysis<sup>[5]</sup>, marking the great potential of LIBS. However, on the way of fully commercialization and industrialization, LIBS is facing three big challenges: 1) to improve the quantitative analysis performance, particularly the repeatability and reproducibility performance; 2) to reduce the instrumental cost; 3) to improve the long-term stability and robustness for industrial applications. To finally transform LIBS from “future superstar” to “superstar”, joint effort of worldwide LIBS community is needed.<sup>[6]</sup>

Currently there are three major LIBS communities promoting LIBS technique, namely, the Euro-Mediterranean LIBS society (EMS-LIBS), the North American LIBS society (NAS-LIBS) and the Asian LIBS society (AS-LIBS). Among the three major LIBS communities, the AS-LIBS community is the youngest one but with the fastest growth both in its scale and influence<sup>[7-1]</sup>. Prior to the first AS-LIBS symposium in 2015, the LIBS community in Asia had already been active from the first Chinese symposium on LIBS (CS-LIBS) successfully held in Qingdao in 2011. The birth of CS-LIBS in 2011 laid the foundation of the Asian LIBS community. Notably, in September, 2014, the 8<sup>th</sup> International Conference of LIBS (LIBS2014) was successfully held in Tsinghua University, China, marking the AS-LIBS community as one of the major communities in the world. Following the success of LIBS 2014, the first AS-LIBS symposium hosted in Wuhan, China in 2015 also achieved a great success with more than 200 attendees<sup>[7]</sup> and set up a milestone for the development of AS-LIBS community. No doubt, it has been regarded as almost the biggest LIBS community among the three major communities in the world. Blessed by the current prosperity of Asian LIBS community, the 2<sup>nd</sup> AS-LIBS symposium was held in Tokushima, Japan in 2017. During the AS-LIBS 2017 symposium, many aspects of the latest LIBS progress were presented in the categories of fundamentals, novel applications, experimental and data processing methods etc. More excitingly, the 11<sup>th</sup> International Conference on LIBS (LIBS 2020) will be hosted in Kyoto, Japan. As the

biggest event in LIBS community is about to return to Asia, AS-LIBS continues to consolidate its important position in the worldwide LIBS community and gives more and more contributions on the development and maturity of the LIBS technique. Therefore, a special issue will be published on “Plasma Science & Technology” to list part of the progress presented on the AS-LIBS 2017 symposium.

There are 22 research articles included in this special issue, providing the readers a rough idea of the very recent achievement of AS-LIBS community. Taking a glimpse of these 22 research articles, a wide range of topics were covered including the fundamentals, experimental and data processing methods and all kinds of applications. Among all the papers, the work on fundamental LIBS is relatively less prominent as the end user demand for quantitative analysis has pushed the LIBS community to put more focus on the experimental, data processing and applications study. Still, important work on fundamental LIBS such as femtosecond dual-pulse LIBS on the signal enhancement<sup>[8]</sup>, pressure effects on underwater LIBS in ocean environment<sup>[9]</sup>, accuracy improvement on CF-LIBS<sup>[10]</sup>, self-absorption effect at ungated scheme<sup>[11]</sup> as well as a review on the mechanism and elimination of self-absorption effect<sup>[12]</sup> was included in this special issue. Several works on the experimental point of view to optimize the LIBS performance were also included, such as a method of laser focusing control in micro LIBS<sup>[13]</sup>, the effect on lens-to-sample distance<sup>[14]</sup>, experimental parameter settings on the matrix effect<sup>[15]</sup> and experimental factors affecting the limit of detection (LOD)<sup>[16]</sup> etc. From the practical point of view, the end users market of LIBS is huge in Asia, leading to a significant focus on the research of applications on LIBS, with a special focus on the data processing methods. This special issue includes a great extent of novel applications of data processing methods on the latest LIBS applications in Asia. Machine learning methods such as SVM<sup>[17, 18]</sup>, K-means<sup>[18]</sup>, PLS-based approaches<sup>[19]</sup>, co-training regression model<sup>[20]</sup>, random forest algorithm<sup>[21]</sup>, GA-KELM method<sup>[22]</sup>, convolution neural networks<sup>[23]</sup> etc were applied to various applications. Methods such as revised PCA categorizing<sup>[24]</sup>, automatic line selection and calibration curve construction<sup>[25]</sup> were also proposed. The versatility of data processing methods has greatly enhanced the quantitative performance of LIBS applications. This special issue also includes many novel applications of LIBS in the AS-LIBS community, such as portable fiber-optic LIBS system on steel analysis<sup>[26]</sup>, high repetition rate LA-SIBS application<sup>[27]</sup>, application on cement raw material<sup>[17]</sup>, heavy metals in water<sup>[28]</sup>, remote LIBS analysis on steel<sup>[29]</sup>, coal and biomass analysis<sup>[18]</sup> etc. As said before, the work included in this special issue partially represents the prosperous research future of LIBS in Asia, there were even more work not included in the special issue.

Summarizing the current research progress from AS-LIBS community not only listed in the special issue but also other journals, a few conclusions and perspectives can be made. First of all, despite the great versatility of LIBS on different applications, it is more and more clear that LIBS should never be considered as a catholic analytical method. In terms of quantitative analysis, it will be very difficult for LIBS to achieve comparable sensitivity and accuracy results from other spectroscopy techniques such as tunable diode laser absorption spectroscopy (TDLAS) and inductively coupled plasma optical emission spectroscopy (ICP-OES). LIBS must maximize the unique advantage based on its ability of performing online and in-situ analysis. Taking the “Curiosity” mars rover from NASA as an example<sup>[30]</sup>, LIBS offers a great convenience for remote in-situ qualitative and semi-quantitative elemental analysis, particularly at extreme environment such as outer space detection. That is, taking advantage of the ability of performing fast online and

in-situ analysis, LIBS should have a great future on the applications of process industry, mineral exploration and extreme environment detection, which is the key pushing force for AS-LIBS to grow from a big LIBS community to a strong LIBS community since of the large market existed in Asia especially in China. To date, AS-LIBS community has made tremendous progress in the aforementioned fields. Several groups including Tsinghua University<sup>[31-38]</sup>, South China University of Technology<sup>[39-42]</sup>, Shanxi University<sup>[43-46]</sup> and Mitsubishi Heavy Industries<sup>[47, 48]</sup> have made a great contribution on the coal property analysis; groups such as Shenyang Institute of Automation<sup>[49-51]</sup>, Huazhong University of Science and Technology<sup>[52, 53]</sup>, and Tohoku University<sup>[54, 55]</sup> also made significant progress on the online and in-situ LIBS applications on the metallurgical industry; In terms of applications for the extreme environment detection, groups such as University of Tokyo<sup>[56-59]</sup> and Ocean University of China<sup>[60-63]</sup> have successfully employed the in-situ analysis of deep-sea mineral samples using LIBS, and groups such as Dalian University of Technology<sup>[64-67]</sup> and Japan Atomic Energy Agency<sup>[68, 69]</sup> have proved that LIBS is an effective tool for in-situ elemental analysis for nuclear reactors. A new collinear DP-LIBS method has been proposed by Tokushima University and Xi'an Jiaotong University. The external energy was supplied by the long pulse-width laser to maintain the plasma to improve the detection ability in the various real applications using its collinear configuration<sup>[70-1, 71-1]</sup>. Another important conclusion is that driven from the application requirement with large application market, AS-LIBS community also make very good progress on the fundamental study and data processing methods. AS-LIBS is currently playing a competitive role on the fundamental studies including various signal enhancement or sensitivity enhancement methods such as magnetic confinement<sup>[70-72]</sup>, spatial confinement<sup>[73-75]</sup>, fast pulse discharge<sup>[76, 77]</sup>, and PLEAF<sup>[78-80]</sup>. Besides, research on improving the signal repeatability with cavity confinement<sup>[81-83]</sup> and the optimization of other experimental parameters<sup>[84, 85]</sup> has also drawn much attention among LIBS community. In terms of data processing methods, contributed by Tsinghua University<sup>[86-90]</sup>, Shenyang Institute of Automation<sup>[91-94]</sup> and many other groups<sup>[95-97]</sup>, AS-LIBS is also playing a leading position of realizing precise quantitative analysis of LIBS. As we can see, AS-LIBS is currently playing a leading role in the LIBS applications in process industry, metallurgical industry and extreme environment detection. AS-LIBS also has great strength in the fundamental study and data processing methods development. Moreover, driven by the massive end users market in Asia, AS-LIBS will continue to strengthen itself in large-scale industrialization of LIBS. It will be an inevitable trend for AS-LIBS to develop from big to strong with the joint effort of all fellow researchers in the AS-LIBS community.

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