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Lead-free piezoceramics − Where to move on?

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Abstract

Lead-free piezoceramics aiming at replacing the market-dominant lead-based ones have been extensively searched for more than a decade worldwide. Some noteworthy outcomes such as the advent of commercial products for certain applications have been reported, but the goal, *i.e.*, the invention of a lead-free piezocermic, the performance of which is equivalent or even superior to that of PZT-based piezoceramics, does not seem to be fulfilled yet. Nevertheless, the academic effort already seems to be culminated, waiting for a guideline to a future research direction. We believe that a driving force for a restoration of this research field needs to be found elsewhere, for example, intimate collaborations with related industries. For this to be effectively realized, it would be helpful for academic side to understand the interests and demands of the industry side as well as to provide the industry with new scientific insights that would eventually lead to new applications. Therefore, this review covers some of the issues that are to be studied further and deeper, so-to-speak, lessons from the history of piezoceramics, and some technical issues that could be useful in better understanding the industry demands. As well, the efforts made in the industry side will be briefly introduced for the academic people to catch up with the recent trends and to be guided for setting up their future research direction effectively.

Keywords: Lead-free piezoceramics; Doping effect; Incipient piezoelectricity; Piezoelectric applications

Contents

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1. Introduction

More than 10 years of both extensive and intensive research activities have been devoted to developing lead-free piezoelectrics in response to the call for environmental protections [1−5]. From the beginning, the goal was obviously to develop lead-free piezoceramics whose properties are no less than those of the market-dominating lead zirconate titanate (PZT). At least when textured potassium sodium niobate (KNN)-based piezoceramics were reported by Saito et al. [6] in 2004, the goal appeared to be achievable quite readily [7]. Although most conceivable material systems have been explored and various strategies have been applied thereafter, the outcome does not seem so exciting as initially expected, given that lead-free piezoceramics are not yet the choice of materials for commercial products, though lead-free piezoceramics are introduced to selected commercial products [8]. Nevertheless, this does not mean that all the collective works by the community members are not noteworthy. In fact, a decade of worldwide research has led to the discovery of a number of potentially promising lead-free piezoceramics as well as valuable contributions to our better understanding of various phenomena in piezoceramics [7,9−21]. However, it is hard for one to deny the fact that scientific research activities are culminated [8], waiting for new issues. Hence, it is right time that we search for how to reactivate scientific research activities both inside and outside the community.

It is said that the relatively long history of piezoelectric community is indebted to the advent of several ground-breaking events such as the invention of $BaTiO_3$ [22] and $Pb(Zr,Ti)O_3$ (PZT) [23], the discovery of morphotropic phase boundary (MPB) [24−27], advanced understandings of relaxor ferroelectrics [28], doping effect based on the defect chemistry [29], and the utilization of polarization rotation in the relaxor-based single crystals [30,31]. As noted, each event has dedicated significantly to the growth of the community both in size and in quality, making the working concepts for developing new materials and applications. It is undeniable that the strategies and approaches taken for developing new lead-free piezoelectric materials are mostly rooted from them. However, it turns out that those concepts are not so efficient for lead-free piezoelectric materials as for lead-containing ones [9−17] Though not clear yet, two reasons could be considered for the unsatisfactory results during the transfer of those working concepts to developing lead-free piezoelectric materials. One possibility is that we might have missed some crucial considerations in establishing those working concepts, *e.g.*, the role of lead ion. Given that the MPB and the polarization rotation concepts have been developed inductively from the experimental observations in the lead-based piezoelectric materials [30,32], we cannot say that the applicability of the concepts onto the other material systems is guaranteed. In fact, there are a great amount of experimental results, the explanation of which does not fit into the classical theory. For example, many of MPBs identified in lead-free piezoceramics did not provide noticeable enhancements in piezoelectric properties like the conventional lead-based piezoceramics [33−40]. As well, the well-defined so-called ‘soft’ and ‘hard’ effects due to the presence of donors and acceptors, respectively, do not seem to be obvious in lead-free piezoceramics, especially the $(Bi_{1/2}Na_{1/2})TiO_3$ (BNT)-based ones [10,41−43]. For example, as will be discussed in a later section, none of the effects due to the presence of either acceptor or donor seems obvious in a number of lead-free piezoceramics in that the soft and hard effect represented by the increase in the electromechanical coupling factor and the mechanical quality factor, respectively, are, in principle, countereffective [44−48].

Apart from the scientific issues, scientific activities can also be significantly boosted up by bringing what has been achieved in the academia to the industry through technology transfers. Efforts in commercialization are sure to introduce new challenges, the solution of which requires further scientific insights. It is of course that scientific research activities to overcome these challenges will give rise to new applications for the industry. This means that the longevity and prosperity of the research field depend greatly on the establishment of this virtuous cycle leading to mutual benefits for both the scientific community and the industry. Here, one of the most important prerequisites is a continuous communication between academia and industry. We believe that this communication needs initiating from the academic side, because the motivation for the transition to lead-free piezoelectrics is rather weak on the industry side.