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**Datasheet for the decision
of 10 December 2019**

Case Number: T 2029/15 - 3.4.03

Application Number: 09816590.5

Publication Number: 2340574

IPC: H01L41/187, C04B35/475,
H01L41/43

Language of the proceedings: EN

Title of invention:

NBT BASED LEAD-FREE PIEZOELECTRIC MATERIALS FOR HIGH POWER
APPLICATIONS

Applicant:

THE PENN STATE RESEARCH FOUNDATION

Headword:

Relevant legal provisions:

EPC Art. 56
RPBA Art. 13

Keyword:

Late-filed main request - admitted (yes)
Inventive step - all requests - (no)

Decisions cited:

Catchword:



Beschwerdekammern
Boards of Appeal
Chambres de recours

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Case Number: T 2029/15 - 3.4.03

D E C I S I O N
of Technical Board of Appeal 3.4.03
of 10 December 2019

Appellant: THE PENN STATE RESEARCH FOUNDATION
(Applicant) The Pennsylvania State University
304 Old Main
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Representative: Ettmayr, Andreas
KEHL, ASCHERL, LIEBHOFF & ETTMAYR
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted on 23 April 2015
refusing European patent application No.
09816590.5 pursuant to Article 97(2) EPC.

Composition of the Board:

Chairman G. Eliasson
Members: M. Papastefanou
W. Van der Eijk

Summary of Facts and Submissions

I. The appeal concerns the decision of the examining division refusing the European patent application No. 09 816 590.5 (published as WO 2010/036363 A2) on the ground that the sole request before it did not meet the requirements of Article 123(2) EPC.

II. At the oral proceedings before the board, the appellant (applicant) requested that the decision under appeal be set aside and that a patent be granted on the basis of the Main Request or one of Auxiliary Requests 1 and 2, all filed with the appellant's letter of 5 December 2019.

III. Reference is made to the following documents:

D1: S.H. Choy et al., *"Electromechanical and ferroelectric properties of $(Bi_{1/2}Na_{1/2})TiO_3-(Bi_{1/2}K_{1/2})TiO_3-(Bi_{1/2}Li_{1/2})TiO_3-BaTiO_3$ lead-free piezoelectric ceramics for accelerometer application"*, Applied Physics A89, 775-781 (2007);

D3: JP 2002-321976 A;

D3a: Automatic English translation of D3, transmitted to the appellant with the communication of the board dated 13 June 2019.

IV. Claim 1 of the Main Request is worded as follows:

A piezoelectric compound having the formula $(xNa_mBi_nTiO_3-yK_mBi_nTiO_3-zLi_mBi_nTiO_3-pBaTiO_3)-rM$ where $(0 < x < 1)$, $(0 < y < 1)$, $(0 < z < 1)$, $(0 < p < 1)$, $(x+y+z+p=1)$, $(0.3 \leq m \leq 0.7)$, $(0.3 \leq n \leq 0.7)$, $(0.9 \leq m/n \leq 1.1)$ and $(0 \text{ wt}\% < r \leq 5 \text{ wt}\%)$ where r is based on the weight of a compound within the scope of $xNa_mBi_nTiO_3-yK_mBi_nTiO_3-zLi_mBi_nTiO_3-pBaTiO_3$ and M is a dopant selected from the

group consisting of Al_2O_3 , CoO , Co_2O_3 , Re_2O_3 where Re is rare earth element, $NiCO_3$, MnO_2 , $MnCO_3$, Fe_2O_3 and mixtures thereof.

- V. Claim 1 of Auxiliary Request 1 has the same wording as claim 1 of the Main Request with the difference that the range of values for x is $0 < x \leq 0.85$.
- VI. Claim 1 of Auxiliary Request 2 has the same wording as claim 1 of the Main Request with the difference that the range of values for x is $0.3 \leq x \leq 0.8$.
- VII. The appellant's arguments could be summarised as follows:

Regarding the admissibility of the newly filed requests, the appellant pointed out that despite being filed only a few days before the oral proceedings, the claims were not new in the procedure. The Main Request filed on 5 December 2019 corresponded essentially to Auxiliary Request 4 filed with the grounds of appeal. The claims of the Auxiliary Requests were similar to the claims of the Main Request. The independent claim of the Main Request had already been subject to a preliminary opinion by the board. Moreover, the new requests addressed most of the objections raised in the board's preliminary opinion, simplified the claimed subject-matter, did not raise any new issues but rather reduced the number of issues to be discussed. The requests should therefore be admitted into the proceedings.

With respect to inventive step, the appellant's main argument was that the skilled person starting from D3 would not be motivated to add Li to the piezoelectric ceramic. According to D1, adding Li to the

piezoelectric ceramic provoked significant modifications to the crystalline structure of the ceramic and the skilled person would actually be taught away from adding Li to the piezoelectric ceramic of D3.

Reasons for the Decision

1. The claimed invention

1.1 The most common piezoelectric materials are the so-called PZT ($\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$) - lead zirconate titanate) materials. Due to the presence of lead (Pb), which is a toxic material, their use has increasingly been considered as hazardous to the environment and efforts have been made to find piezoelectric materials which do not contain any lead and perform as well as the PZT materials.

A known family of such lead-free piezoelectric materials is based on the compound $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ ("NBT" or "BNT") in combination with further components based on potassium (K), lithium (Li), and barium (Ba). These materials, however, have disadvantages when compared to the PZT piezoelectric materials, such as low T_c (transition/Curie temperature), low piezoelectric activity, multiple phase transitions etc. (see page 1, line 11 to page 2, line 16 of the published application).

1.2 The invention addresses the problem of providing lead-free piezoelectric materials that overcome the disadvantages of the lead-free piezoelectric materials of the prior art (page 2, line 17 to page 3, line 1).

The claims define such piezoelectric materials (and

methods for their manufacture) based on the general formula $x\text{Na}_m\text{Bi}_n\text{TiO}_3 - y\text{K}_m\text{Bi}_n\text{TiO}_3 - z\text{Li}_m\text{Bi}_n\text{TiO}_3 - p\text{BaTiO}_3$ (NBT-KBT-LBT-BT), with specific value ranges for the parameters x , y , z , m , n and p and the possible use of additional acceptor dopants (see page 4, line 20 to page 5, line 2). The addition of metallic dopants (see "M" in the formula of the claims 1) further improves the quality of the piezoelectric ceramic.

2. Admissibility of the newly filed requests

2.1 The Main Request as well as Auxiliary Requests 1 and 2 were filed on 5 December 2019, only a few days before the oral proceedings before the board.

2.2 The board agrees with the appellant that the requests do not contain any subject-matter that was filed for the first time. The Main Request in particular corresponds essentially to Auxiliary Request 4 filed with the grounds of appeal. Claim 1 of the new Main Request is the same as Claim 1 of Auxiliary Request 3 filed with the statement of the grounds of appeal.

2.3 Regarding the Main Request, it comprises only one independent claim per category and overcomes all the objections under Articles 84 and 123(2) EPC raised in the board's preliminary opinion without raising any new issues. Having considered claim 1 in the context of its preliminary opinion (see point 6.3 of the board's communication of 13 June 2019, relating to claim 1 of Auxiliary Request 3), the board could deal with the Main Request without adjourning the oral proceedings (Article 13(3) Rules of Procedure of the Boards of Appeal (RPBA 2007)).

Hence, the board, exercising its discretion under

Article 13(1) RPBA 2007 decided to admit the Main Request into the proceedings.

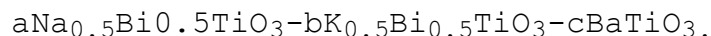
2.4 Regarding the Auxiliary Requests, the board did not consider it necessary to decide on their admissibility as the board considered them not to be allowable, as will be explained in points 4 and 5.

3. Main Request

3.1 Closest prior art

Document D3 addresses the same technical problem as the present application, namely to provide a piezoelectric ceramic which has similar properties as the known PZT piezoelectric ceramics but contains no lead (see paragraphs [0002] to [0005] in D3a). D3/D3a describes a piezoelectric material with the general formula $\text{NaBiTiO}_3\text{-KBiTiO}_3\text{-BaTiO}_3$ (NBT-KBT-BT) (see Abstract or Table 1 of D3, for example).

More specifically the compound described in D3 has the formula:



The corresponding parameters from the formula have the following value ranges (see abstract of D3):

- $0.45 \leq a \leq 0.99$ (corresponding to "x" in claim 1)
- $0 < b \leq 0.35$ (corresponding to "y" in claim 1)
- $0 < c \leq 0.2$ (corresponding to "p" in claim 1)
- "m"=0.5
- "n"=0.5
- there is no LBT, so "z"=0.

The piezoelectric material of D3 comprises also a dopant of the same type and in the same quantity/ proportion as in claim 1 (5 wt% or less - corresponding to the "r" of claim 1) (see D3a, paragraphs [0034] to

[0039]).

Since D3/D3a achieves the same purpose and comprises the most technical features in common with the claimed invention, it is considered to represent the closest prior art.

3.2 Difference and technical problem

3.2.1 The only feature distinguishing claim 1 of the Main Request from D3 is that the claimed piezoelectric compound comprises a lithium - based component (LiBiTiO₃ - LBT).

3.2.2 The appellant pointed out that piezoelectric ceramics are complicated compounds with crystalline structures. The presence or absence of Li (or LBT) from the piezoelectric compound implied different crystalline structures of the compounds and this should also be taken into account when considering the differences between the claimed compound and the compound of D3.

3.2.3 Making reference to Tables II and IV of the published application, the appellant identified the different technical effects of the two compounds. In the first line of Table II (page 21 of the published application) measurements of various properties of the piezoelectric material of Example 1G were given (see also page 17, lines 31 to 34). The compound of Example 1G had the general formula NBT-KBT-BT-rM, which corresponded to the compound of D3. In the last three lines of Table IV (page 22) measurements of the same properties of the piezoelectric compounds of Examples 5A, 9 and 10 were given (see page 19, lines 9 to 16 and page 20, lines 8 to 23). These materials had the general formula NBT-KBT-LBT-BT-rM, which corresponded to the compound of

claim 1. From the two tables, it was clear that the properties d_{33} (piezoelectric charge constant) and Pr (remnant polarization) were "significantly improved" in the compound containing LBT (i.e. Li) with respect to the compound without LBT.

3.2.4 Starting from D3, the skilled person would thus be faced with the objective technical problem of how to improve piezoelectric charge constant (d_{33}) and the remnant polarization (Pr) of the BNT-KBT-BT-rM piezoelectric compound.

3.2.5 The board decided to follow the appellant in the formulation of this objective technical problem.

3.3 Solution and obviousness

3.3.1 The claimed piezoelectric compound solves the identified technical problems with the addition of an LBT (Li-based) compound. As Tables II and IV show, the values of these two properties are improved with the addition of LBT, hence the claimed compound solves the identified objective technical problem.

3.3.2 In the board's view, the skilled person seeking to improve the two identified properties (d_{33} and Pr) of the piezoelectric compound of D3 would consider document D1.

D1 is a scientific publication which studies the advantages of using lithium (LBT) in an NBT-KBT-BT piezoelectric compound (see page 776, left column, first paragraph). As can be seen in the title of D1, the described piezoelectric compound comprises a lithium based component (LBT) and corresponds to the piezoelectric compound of claim 1 of the Main Request,

without the dopant (see also D1, page 776, left column under the title "**2 Experimental procedures**").

3.3.3 In the board's view, the individual components of the various piezoelectric materials described in the prior art documents and the application (NBT, KBT, LBT, BT and the additional dopants) were known to the skilled person before the priority date of the application. This seems also to be indicated in the application itself (see section "**BACKGROUND OF THE INVENTION**" starting on page 1 of the published application). In the context of the claimed invention, the quest for an improved lead-free piezoelectric material seems to relate to finding an appropriate combination of (some of) these individual components so that the piezoelectric performance of the material produced by this combination is considered satisfactory with respect to PZT piezoelectric materials. This appears to be corroborated by the examples in the application where piezoelectric materials consisting of (some of) these components (NBT, KBT, LBT, BT and other dopants) in various combinations are produced (see page 15, line 17 to page 20, line 23).

The same is also indicated both in D3 (see paragraphs [0006] to [0008] of D3a) and in D1 (see section "**1 Introduction**"). It appears, therefore, that the skilled person would select some of these known combinations to try to manufacture a satisfactory piezoelectric compound. Neither the prior art documents nor the application provides any indication that a specific combination of these components would not be possible. Rather, the corresponding advantages and disadvantages of different combinations are discussed. The possibility of adding metallic dopants to these combinations is also discussed. From these discussions

the board concludes that the presence of a metallic dopant (such as an oxide of Al, Co or Mn for example) is not dependent on specific combinations of the NBT, LBT, KBT and BT compounds. In other words, there is no indication that the addition of a specific component (e.g. LBT) in the piezoelectric compound would be incompatible with the use of a metallic dopant.

Moreover, D3 provides an indication that other elements may be added to the main piezoelectric compound described (see paragraph [0074] of D3a). At this point the board agrees with the appellant that a selection of Li from the list of the elements mentioned in this paragraph of D3a would not be obvious for a skilled person based on the disclosure of D3/D3a. However, this passage is referenced only as an indication that, in the context of D3/D3a, the addition of other elements to the described piezoelectric compound is not excluded but rather suggested.

- 3.3.4 Regarding the effects of the addition of an LBT compound in the NBT-KBT-BT (or BNKBT in D1) piezoelectric material, it is explicitly mentioned in D1 that the incorporation of a "proper amount" of LBT into the NBT-KBT-BT compound, provides for a "significantly larger" remnant polarization (P_r) (see the paragraph bridging the two columns on page 777 and the inserted diagram in Figure 4 on page 778).
- 3.3.5 With respect to the piezoelectric discharge constant (d_{33}), D1 states that the compound including LBT has a higher d_{33} constant in comparison to the compound without LBT (see bottom of left column on page 778).

The appellant pointed out that according to the bottom diagram of Figure 5 (page 778) the value of d_{33}

decreased as the quantity of LBT in the compound increased, with the value being higher when there was no LBT at all (amount of LBT 0.0 mol%). This contradicted the statement regarding the increase of d_{33} with the addition of LBT referred to by the board. Therefore, the skilled person would not derive directly and unambiguously from D1 that the addition of LBT in the piezoelectric compound improved the values of d_{33} .

Although the board acknowledges this inconsistency regarding the effects of adding LBT into the NBT-KBT-BT compound in D1, it is of the opinion that this would not stop the skilled person from applying the teaching of D1 in D3. The skilled person reading D1 would learn that adding LBT into the compound would clearly improve the remnant polarization (P_r), i.e. one of the two properties he is seeking to improve. He would also get a hint that an improvement of piezoelectric discharge constant (d_{33}) may also be possible, although there would remain some doubts in view of the diagram in Figure 5. He would also learn that the addition of a proper amount of LBT would improve a number of other properties of the piezoelectric material such as the electromechanical coupling factors, the mechanical quality factors and the transition temperature (T_c) (see bottom of left column and the first two paragraphs of the right column on page 778). He would thus apply the teaching of D1 and add an LBT compound into the material of D3, obtaining thus the claimed compound in an obvious and straightforward manner.

- 3.3.6 The appellant argued that D1 taught that adding Li to the NBT-KBT-BT compound changed its crystalline structure from rhombohedral to tetragonal (see abstract and penultimate paragraph of the left column on page 777). The skilled person would thus be taught away from

adding Li to the piezoelectric compound of D3 because he knew that it would modify its crystalline structure with unforeseen consequences to its various properties and overall performance.

3.3.7 The board does not agree with the appellant. According to D1, the incorporation of Li in the NBT-KBT-BT compound improved a series of relevant properties (and overall piezoelectric performance) despite the modification caused in its crystalline structure. There was no suggestion in D1 that the change of crystalline structure caused any deterioration to the material. In addition, the piezoelectric compound in D3 has both rhombodendral and tetragonal crystalline structures (see D3a, paragraph [0013]). In the board's view, the skilled person would not be discouraged from applying the teaching of D1 to D3.

3.4 The conclusion of the board is that the subject-matter of claim 1 of the Main Request does not involve an inventive step within the meaning of Article 56 EPC.

4. Auxiliary Request 1

4.1 The board notes that the range of the parameter "a" in D3 (0.45 to 0.99), which corresponds to the "x" of the claim, is overlapping with the value range of "x" in claim 1 of Auxiliary Request 1 (0 to 0.85). Therefore, the features distinguishing claim 1 of Auxiliary Request 1 from D3 are the same as those distinguishing claim 1 of the Main Request from D3. The reasoning and the conclusion regarding inventive step are thus the same for Auxiliary Request 1 as for the Main Request.

4.2 The appellant noted that in the piezoelectric compound of D1, which contained LBT, the amount of the NBT

compound was limited to 0,875 to 0.9 (the parameter corresponding to the "x" of claim 1, see first lines under "**2. Experimental procedures**", on page 776)). Even if in D3 the amount of NBT could take values in the range of 0.45 to 0.99, the skilled person would learn from D1 that in a compound comprising LBT, the amount of NBT would be limited to values between 0.875 and 0.9, which was within the range defined in D3 but outside the range of claim 1 of Auxiliary Request 1.

Therefore, the skilled person seeking to solve the same objective technical problem as identified for the Main Request, and even if he were to combine D3 and D1, he would still not arrive at the claimed product since the value range for "x" would not be the same. The subject-matter of claim 1 of Auxiliary Request 1 was thus inventive.

4.3 The board does not share the appellant's opinion.

On one hand, as the appellant argued, the selection of a value range for x of 0 to 0.85 does not provide any particular technical effect with respect to the value range of 0 to 1, since no particular technical problem was addressed by this selection. This leads to the conclusion that the specific value range is an arbitrary selection without any inventive merits.

On the other hand, D1 is a scientific paper studying the effects of adding various amounts of LBT to a piezoelectric compound of the type NBT-KBT-BT of a particular composition (see ABSTRACT). It would be expected, thus, that in the context of D1 the initial amounts of the various components (NBT, KBT, BT) would be varied as little as possible so that the effects of LBT on the resulting compound could be studied. In the

board's view, the skilled person would not derive from the teaching of D1 that the identified improvements in the various properties of the piezoelectric compound were achieved only with the specific amounts of the individual compounds mentioned.

Moreover, the product of D3/D3a was the starting point for the skilled person and not the product of D1. In D3, the value range of x ("a" in D3/D3a) is broader than in D1 (0.45 to 0.99 in D3 compared to 0.875 to 0.9 in D1) and the skilled person adding the lithium-based compound (LBT) in the product of D3 would hence arrive at the claimed compound in an obvious and straightforward manner.

4.4 The board's conclusion is, therefore, that the subject-matter of claim 1 of Auxiliary Request 1 does not involve an inventive step.

5. Auxiliary Request 2

5.1 The board notes that, as with the other requests, the range for "x" in claim 1 of Auxiliary Request 2 ($0.3 \leq x \leq 0.8$) is not new with respect to D3, which discloses a range of 0.45 to 0.99 for the corresponding parameter.

Since this feature is disclosed in D3, the only feature distinguishing claim 1 of Auxiliary Request 2 from D3 is the LBT compound, as in the Main Request. Therefore, the subject-matter of claim 1 of Auxiliary Request 2 does not involve an inventive step for the same reasons as claim 1 of the Main Request.

5.2 The appellant pointed out that the range of x in claim 1 of Auxiliary Request 2 was clearly outside the

corresponding range in D1 (0.3 to 0.8 in the claim, 0.875 to 0.9 in D1).

Regarding the technical problem solved by this particular value range, the appellant referred to Example 10 of the application (see page 20, lines 17 to 19). As it could be seen in the last line of Table IV (page 22), Example 10 "significantly improved" values for d_{33} and P_r compared to the compound without LBT (see first row of Table II, page 21) and achieved the "best trade-off" regarding the remaining properties when compared to Examples 5A and 9 (see the three last rows on Table IV). The objective technical problem in this case would be therefore how to improve d_{33} and P_r while obtaining the best trade-off with respect to the values of the other properties of the piezoelectric compound. The skilled person combining D3 and D1 would not arrive at the claimed piezoelectric compound because in D1 the corresponding x parameter was taking values outside the range of claim 1 of Auxiliary Request 2.

- 5.3 The board does not agree with the argument of the appellant.
- 5.3.1 At first, as explained also with respect to Auxiliary Request 1, the value range of x in D1 is not relevant since the starting product for the skilled person is the piezoelectric compound of D3 in which the corresponding value range overlaps with the one of claim 1 (see point 4.3 above).
- 5.3.2 Secondly, comparing the values of the various parameters in Examples 5A, 9 and 10 one can see that the values of several parameters (x , y , z , p) are modified from one example to the other. It cannot be

asserted, therefore, that any particular technical advantage obtained in Example 10 is the result of the specific value of x (0.8) only.

- 5.3.3 Moreover, even if this were the case, there is nothing in Example 10 or the application as a whole that would support the appellant's argument that the identified technical effect is achievable for the value range of x as defined in claim 1 of Auxiliary Request 2 (0.3 to 0.8). Example 10 provides specific measurements of specific properties for specific values of the various parameters of the formula. There is nothing suggesting that the amounts could be generalised to the amounts of the ranges of the claim.
- 5.3.4 The board's opinion is therefore that the particular value range for the parameter " x " in claim 1 of Auxiliary Request 2 does not address any particular technical problem or provide any particular technical effect. It is rather an arbitrary selection from the range in claim 1 of the Main Request (0 to 1), which cannot provide a basis for an inventive step within the meaning of Article 56.
- 5.4 The board concludes, hence, that the subject-matter of claim 1 of Auxiliary Request 2 does not involve an inventive step for the same reasons as for claim 1 of the Main Request.
6. Since none of the requests on file is allowable, the appeal must fail.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:



S. Sánchez Chiquero

G. Eliasson

Decision electronically authenticated