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**Datasheet for the decision
of 8 February 2018**

Case Number: T 1587/14 - 3.3.03

Application Number: 06014918.4

Publication Number: 1754736

IPC: C08G61/12

Language of the proceedings: EN

Title of invention:

Process for the polymerisation of thiophene or selenophene derivatives

Patent Proprietor:

Merck Patent GmbH

Opponent:

BASF SE

Headword:

Relevant legal provisions:

EPC Art. 56

RPBA Art. 13(3)

Keyword:

Inventive step - (no) Main request, first and second auxiliary requests - two unrelated partial problems
Late-filed auxiliary requests - admitted (no) third to fifth auxiliary requests

Decisions cited:

Catchword:



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Case Number: T 1587/14 - 3.3.03

D E C I S I O N
of Technical Board of Appeal 3.3.03
of 8 February 2018

Appellant: BASF SE
(Opponent) 67056 Ludwigshafen (DE)

Representative: BASF IP Association
BASF SE
G-FLP-C006
67056 Ludwigshafen (DE)

Respondent: Merck Patent GmbH
(Patent Proprietor) Frankfurter Strasse 250
64293 Darmstadt (DE)

Representative: Merck Patent GmbH
64271 Darmstadt (DE)

Decision under appeal: **Interlocutory decision of the Opposition
Division of the European Patent Office posted on
22 May 2014 concerning maintenance of the
European Patent No. 1754736 in amended form.**

Composition of the Board:

Chairman D. Semino
Members: M. C. Gordon
C. Brandt

Summary of Facts and Submissions

- I. The appeal of the opponent lies from the interlocutory decision of the opposition division posted on 22 May 2014 according to which European patent number 1 754 736 could be maintained in amended form on the basis of the main request as filed at the oral proceedings.
- II. The patent was granted with a set of 22 claims, whereby claim 1 read as follows:

"Process for polymerising a heteroaromatic compound under formation of aryl-aryl C-C couplings, wherein said compound has at least one functional halide group and at least one functional boron group, or copolymerising at least one first and at least one second heteroaromatic compound under formation of aryl-aryl C-C couplings, wherein said first compound has at least two functional halide groups and said second compound has at least two functional boron groups, wherein the polymerisation is carried out in presence of

a) a catalyst/ligand system comprising a palladium catalyst and an organic phosphine or phosphonium compound,

b) a base,

c) a solvent or a mixture of solvents,

characterized in that

the functional halide and boron groups are attached to a thiophene or selenophene ring that is optionally substituted and optionally fused to another ring, and the organic phosphine or phosphonium compound is a trisubstituted phosphine or phosphonium salt with substituents selected from optionally substituted alkyl and aryl groups and the organic phosphine compound is

formed *in situ* from the corresponding phosphonium salt during the process by addition of the base."

Claims 2-22 were directed to preferred embodiments of the process.

III. A notice of opposition against the patent was filed in which revocation of the patent on the grounds of Article 100(a) EPC (lack of novelty, lack of inventive step), Article 100(b) EPC and Article 100(c) EPC was requested.

The following documents, *inter alia* were cited in support of the opposition:

D3: WO-A2-2003/048225

D5: Netherton, M.R. *et al*, Organic Letters, 3 (26)
(2001) 4295-4298

In the course of the opposition proceedings both parties submitted additional experimental data:

Patent proprietor: examples 11, 12, 13-17 and comparative examples 13-17 with letter of 19 December 2013;

Opponent: Comparative examples 1 and 2 with letter of 23 October 2013 and comparative examples 3 and 4 with letter of 16 January 2014.

IV. The decision of the opposition division was based on the claims of the main request filed at the oral proceedings on 23 January 2014 (corresponding to auxiliary request 2 filed with letter of 19 December 2013).

Claim 1 of this request differed from claim 1 of the patent as granted by deletion of the feature "or selenophene". Thus the relevant part of the claim read as follows:

"[...] characterized in that the functional halide and boron groups are attached to a thiophene ring that is optionally substituted [...]".

The remaining claims were unchanged.

According to the decision examples 3 and 4 filed by the opponent and examples/comparative examples 13-17 of the patent proprietor were not admitted to the procedure. No explicit decision was given in respect of the additional evidence filed by the parties.

The requirements of Article 123(2) EPC and sufficiency of disclosure were satisfied. Objections pursuant to Article 84 EPC were dismissed since the features objected to had been present in the granted claims. The requirements of novelty were met.

An inventive step was recognised. The available evidence did not demonstrate a technical effect over the closest prior art represented by D3. Thus the objective problem was to provide an alternative process. This was achieved by using different catalysts to D3 (the corresponding phosphonium salts instead of phosphines). D3 did not illustrate the coupling of two thiophene monomers. D5, which disclosed the use of phosphonium salts in Suzuki reactions also did not relate to thiophene polymers, but was concerned with the problems of handling the phosphine components of the catalyst in view of their oxidation sensitivity. D3 in its turn did not address the problem of handling the

phosphine compounds. Thus it would not have been obvious in view of the teaching of D5 to employ phosphonium salts in order to provide an alternative process to that of D3.

- V. The opponent (appellant) appealed against the decision. Together with the statement of grounds of appeal all documents cited in the procedure were resubmitted, as were the experimental data of both parties. Objections pursuant to Article 123(2), 83, 84, 54 and 56 EPC were maintained.
- VI. The patent proprietor (respondent) replied to the statement of grounds of appeal without filing any further claim requests.
- VII. The Board issued a summons to oral proceedings and a communication setting out its preliminary view of the case.
- VIII. With letter of 5 January 2018 the respondent made a further written submission submitting five sets of claims as first to fifth auxiliary requests and a further experimental report (example 18 and comparative examples 18-21).

In the first auxiliary request, compared to the main request the alternative "phosphonium compound" had been deleted from claim 1.

In the second auxiliary request in addition to the amendment as noted for the first auxiliary request, it was specified in part (a) of the claim that the phosphine compound was a "trisubstituted phosphine ligand that was capable of coordinating to the Pd atom" and in the final part of the claim the phosphine moiety

was defined as a "trisubstituted phosphine ligand".

In the third-fifth auxiliary requests in addition to the modifications as noted for the second auxiliary request the definition of the heteroaromatic compounds had been progressively restricted. The details of these restrictions are not of relevance to the present decision.

IX. Oral proceedings were held before the Board on 8 February 2018.

X. The arguments of the appellant insofar as relevant for the present decision can be summarised as follows:

(a) Main request - inventive step

Closest prior art was D3, which related to Suzuki type polymerisation *inter alia* of monomers having thiophene structure. The subject-matter of the main request was distinguished therefrom by the step of *in situ* formation of the phosphine compound from the phosphonium salt.

The experimental data provided by the respondent showed variable, inconsistent effects i.e. unequivocal evidence for a technical effect associated with the manner of introduction of the phosphine component of the catalyst system did not exist.

Consequently the objective problem had to be formulated as the provision of a further process.

From D5 it was known, in order to simplify handling, to carry out Suzuki reactions by adding the

phosphine component in the form of the more stable phosphonium salt and that this gave similar results to addition in the form of the free phosphine. Even though D5 did not relate to polymerisation, the type of phosphine compound envisaged was that employed in D3 for the polymerisation.

(b) First and second auxiliary requests

The arguments in respect of the main request applied also to these requests, no additional counter-arguments being advanced in respect thereof.

(c) Third to fifth auxiliary requests - admittance

The requests had been filed late and there was no justification for this. Furthermore they raised further issues, in particular with respect to Article 123(2) EPC, meaning that they were not clearly allowable and consequently should not be admitted to the procedure.

XI. The arguments of the respondent can be summarised as follows:

(a) Main request - inventive step

It was agreed that D3 was the closest prior art.

D3 did not disclose thiophene polymers. Only a single thiophene (co)monomer was explicitly disclosed. Hence there was an additional difference to that identified by the appellant.

D3 addressed problems associated with the solvent system, not with the catalyst, although it was recognised therein that triphenyl phosphine was not desirable. Thus the aim of D3 was to identify a suitable solvent system and to avoid the use of triphenyl phosphine, this latter problem being solved by use of a substituted phosphine. D3 provided no hint to employ other phosphines or to replace the phosphine by its salt.

D5 related to Suzuki reactions of low molecular weight compounds, but not to polymerisation. The citation of this document was based on *ex post facto* considerations.

The claimed process permitted Suzuki polymerisation of thiophene without the disadvantages of prior art processes known to be associated with polymerisation of thiophene, in particular deboronation. The claimed process also permitted easier handling of the phosphonium salt.

The experimental evidence of examples/comparative examples 13-17 showed an improvement in a large majority of the cases. Specifically the effect of replacing the phosphine by its salt was an increase in molecular weight (three out of five cases) and/or a reduction in polydispersity (four out of five cases).

(b) First and second auxiliary requests

No additional arguments were advanced compared to the main request.

(c) Third to fifth auxiliary requests - admittance

These represented fall-back positions for inventive step in the case that the main request was not accepted.

- XII. The appellant requested that the decision under appeal be set aside and that the European patent No. 1754736 be revoked.
- XIII. The respondent requested that the appeal be dismissed (main request), alternatively, that the patent be maintained in amended form on the basis of one of the sets of claims according to the first to fifth auxiliary requests as filed with letter dated 5 January 2018.

Reasons for the Decision

1. Although both parties, in addition to inventive step, made submission in respect of allowability of amendments, sufficiency of disclosure, clarity and novelty, in view of the conclusions reached with respect to inventive step and admittance of late-filed requests, these latter aspects are not of relevance to the decision and consequently do not need to be addressed.
2. Admittance of late filed evidence to the procedure.

Both parties submitted during the opposition further evidence in the form of examples and comparative examples, which evidence was not admitted by the opposition division. The respondent filed further evidence during the appeal proceedings.

No objections were raised by either of the parties to the various sets of data. On the contrary, both parties relied on said evidence, in particular that provided by the respondent in their submissions, written and oral. Consequently the Board can identify no reason not to admit these data to the procedure.

3. Main request - inventive step

3.1 The patent in suit

The patent is directed to the polymerisation of thiophene derivatives for preparing conjugated thiophene polymers (paragraph [0001], claim 1). According to the section "Background and prior art", in particular paragraph [0005], various methods are known, including the Suzuki reaction. However all of these have disadvantages. In the case of the Suzuki reaction, the problems are low molecular weight and/or low yields, which are attributed to deboration during the reaction (paragraph [0009]).

Hence it is an aim of the patent to provide an improved process for preparing thiophene polymers free from the drawbacks of the prior art methods (paragraph [0013]).

According to paragraph [0016] this problem is solved by a process including as one step the formation of the phosphine ligand from a corresponding salt.

3.2 Closest prior art.

It is a matter of consensus between the parties that the closest prior art is represented by the disclosure of D3.

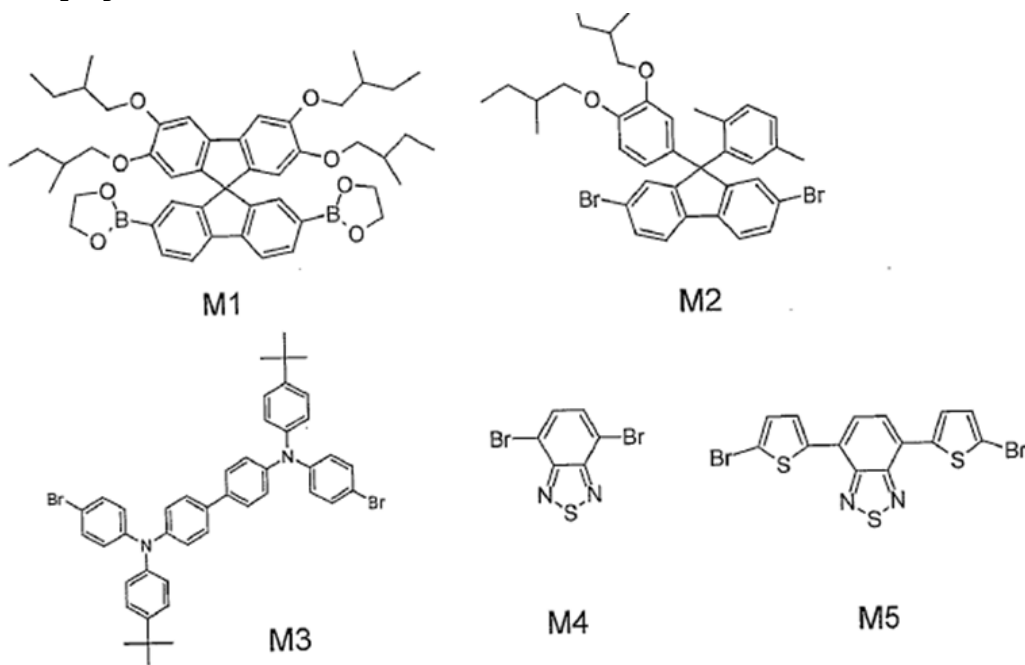
This document relates according to page 1, first paragraph to the preparation of aryl-aryl coupled compounds. It is explained at page 1, second paragraph that there are various problems with such reactions, e.g. mis-reaction and generation of by-products that have to be removed. In the third paragraph of page 1 polymerisation is given as an example of the type of reactions which can be accomplished with the process. On page 2, third paragraph the Suzuki reaction is discussed as a suitable reaction for preparation of aryl-aryl coupled compounds. The following paragraph discusses variations of the Suzuki process. At page 3, first bullet point, problems with use of water soluble palladium-phosphine complexes due to insolubility in non-polar media are mentioned. The second bullet point on page 3 states that the Suzuki reaction is effective in the case of simple molecules but not in the case of polymers due to low yields.

Thus, according to the paragraph bridging pages 5 and 6 the problem which D3 addresses is the provision of an efficient process, at low catalyst concentration, for preparation of aryl-aryl coupled compounds, the solution being the use of particular solvent combinations.

According to page 6, final paragraph and page 7 first paragraph, the aromatic structures employable include thiophenes (page 7, line 4). According to page 7, lines 6-8 polyfunctional compounds can be employed as can oligomers with functional end groups. It is explained at page 7 line 9 to page 8, line 4 that suitable starting materials are substituted or unsubstituted mono- or polyfunctional compounds whereby for producing polymers difunctional compounds are employed. In this passage it is also stated that the starting compounds

can bear either of the functional groups necessary for the Suzuki reaction (boron or halogen). Phosphine ligands are discussed on page 8, the final two lines stating that substituted triphenyl phosphines are particularly preferred. At page 9, 11th line below the figure t-butyl-di-o-tolylphosphine is mentioned as a particularly preferred phosphine.

In D3 only a single example of a thiophene based (co)monomer is shown in Example A2, Polymer P3, monomer M5 (pages 18-20):



In this example following establishment of the monomer solution tris-o-tolyl phosphine and then palladium acetate is added (page 20, example P3).

3.3 Distinguishing features

The subject-matter of operative claim 1 is thus distinguished from the disclosure of D3 by two features:

- both functional monomers involved have a thiophene structure
- the phosphine ligand is generated *in situ* from its salt.

3.4 Technical effect

The examples of the patent itself do not relate to either of these differences and consequently cannot provide evidence for a technical effect. This aspect is not in dispute.

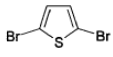
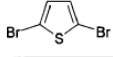
The respondent filed as experimental data, examples/ comparative examples 13-17 during the opposition procedure. the results of which are summarised in the following table:

Table 1: Examples 13-17 and Comparative Examples C13-C17

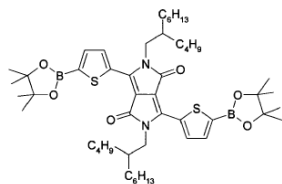
Example	Dibromide	Diboronate	Ligand	Solvent	M _n (kg/mol)	M _w (kg/mol)	PDI	M _n vs. Comp. Ex.	PDI vs. Comp. Ex.
13			[(t-Bu) ₃ PH]BF ₄	THF	27.9	83.5	2.93	+	+
C13			(t-Bu) ₃ P	THF	26.7	108.6	4.07		
14			[(t-Bu) ₃ PH]BF ₄	THF: Toluene (1:1)	23.1	114.2	4.94	+	-
C14			(t-Bu) ₃ P	THF: Toluene (1:1)	18.5	64.2	3.47		
15			[(t-Bu) ₃ PH]BF ₄	THF	14.9	24.9	1.67	+	+
C15			(t-Bu) ₃ P	THF	8.8	18.7	2.13		
16			[(t-Bu) ₃ PH]BF ₄	THF: Toluene (1:1)	20.1	44.8	2.23	-	+
C16			(t-Bu) ₃ P	THF: Toluene (1:1)	23.5	53.2	2.26		
17			[(t-Bu) ₃ PH]BF ₄	THF: Toluene (1:1)	15.0	82.6	5.54	o	+
C17			(t-Bu) ₃ P	THF: Toluene (1:1)	15.4	97.0	6.30		

During the appeal proceedings a further set of data was filed in which example 18 and comparative example 18 are relevant for this decision:

Table 1:

Ex.	Monomer A	Monomer B	Ligand	Ligand	Mon. A (mg)	Mon. B (mg)	THF (cm ³)	Pd cat (mg)	2M K ₂ CO ₃ (cm ³)	Mn (kg/mol)	Mw (kg/mol)	PD
18		see below	[(t-Bu) ₃ PH]BF ₄	6.2 mg	61,7	226,7	5,1	4,7	0,38	39,6	60,1	1,52
C18		see below	(t-Bu) ₃ P	16.7 μl	62,2	228,5	5,1	4,7	0,39	34,7	50,8	1,46

Monomer B:



These data provide a side-by side comparison of adding the phosphine in the form of its salt and in the free form (comparative examples) in otherwise identical processes.

The results can be summarised as follows whereby in the last column "+" denotes an increase in molecular weight or a reduction in polydispersity respectively.

Example pair	Mn	PDI	Result
13/C13	27.9/26.7	2.93/4.07	+/+
14/C14	23.1/18.5	4.94/3.47	+/-
15/C15	14.9/8.8	1.67/2.13	+/+
16/C16	20.1/23.5	2.23/2.26	-/+
17/C17	15.0/15.4	5.54/6.30	-/+
18/C18	39.6/34.7	1.52/1.46	+/-

The evidence of these examples, as acknowledged by the respondent, is that the effect of introducing the phosphine component in the form of its salt is inconsistent. In 4 out of 6 cases an increase in molecular weight is observed and in - a different subset - 4 out of 6 cases a reduction in polydispersity is observed, and whereby in two cases both of these effects are observed simultaneously.

There is no evidence relating to the other difference with regard to D3, namely the structure of the monomer, i.e. whether a thiophene structure or not is used.

3.5 Objective technical problem - its solution

As follows from the above discussion of the available

data, the effect of the *in situ* generation of the phosphine ligand from its salt is inconsistent and no examples exist which are suitable to demonstrate whether any technical effect is associated with the nature of the monomer (thiophene structure). Thus there is also no evidence for a technical effect arising from this feature, and, by corollary no evidence for any effect arising as a result of the two identified distinguishing features in combination.

Under these circumstances the subject-matter claimed has to be seen as relating to an aggregation of distinguishing features with respect to the closest prior art. This in turn means that it will be necessary to adopt the approach of two independent partial problems (see "Case Law of the Boards of Appeal of the European Patent Office", 8th Edition (2016) section I.D.9.2.2).

The first partial problem can be formulated as the provision of a process giving rise to polymers having a different structure to those of D3 and was solved by employing as both functional monomers those having a thiophene structure.

The second partial problem, in the light of the above evidence and the inconsistent results will have to be formulated as the provision of a further process, i.e. devoid of any particular effect or improvement. This problem was solved by employing the phosphine compound in the form of its salt.

3.6 Obviousness

Regarding the first partial problem, as explained above in the discussion of D3, page 7, line 4 thereof explicitly teaches that monomers having thiophene structure can be employed and in the following section states that the two types of functional group (halogen or boron) can be present on any of the aromatic monomer structures disclosed.

Consequently the first partial problem was solved by arbitrarily selecting the nature of the monomer units from among those discussed in D3. An arbitrary restriction to a subset of those structural units disclosed is an obvious and hence non-inventive route to solve the above formulated problem of providing a process resulting in polymers of different structures to those explicitly disclosed in D3.

Regarding the second partial problem, it is known from D5 that trialkyl phosphonium salts provide a "simple, practical and versatile" replacement for air sensitive trialkyl phosphines (title). One of the phosphine compounds envisaged is $[(t\text{-Bu}_3)\text{PH}]\text{BF}_4$ (page 4296, left hand column, third complete paragraph), which is the salt employed in the above examples of the respondent. This phosphine, in the free form, is also mentioned in D3, page 9, 11th line below the figure. According to D5, page 4297, right-hand column, last complete paragraph, this salt is applicable in the Suzuki reaction. It is correct, as argued by the respondent, that D5 is directed to low molecular weight compounds, i.e. not polymers. However the problems which D5 addresses relating to the handling of the phosphine compound are independent of the nature of the starting materials or intended end-product of the reaction, and

consequently apply equally to both low molecular weight and polymer chemistry.

Accordingly with respect to the second partial problem formulated above, i.e. the provision of a further process for carrying out the Suzuki polymerisation, D5 provides a teaching how this could be carried out, i.e. by replacing the free phosphine compound by its salt, rendering the claimed solution to this second partial problem obvious.

3.7 Accordingly the main request does not meet the requirements of Article 56 EPC.

4. First and second auxiliary requests

As explicitly acknowledged by the respondent, the amendments made to these requests were introduced in order to take account of objections pursuant to Article 123(2) EPC, but did not affect the question of inventive step.

Indeed the amendments made do not introduce any additional distinguishing features with respect to closest prior art D3.

Accordingly for the same reasons as given for the main request, these requests are found not to meet the requirements of Article 56 EPC. In view of this there is no need to decide on their admittance.

5. Third to fifth auxiliary requests - admittance to the proceedings

These requests were submitted with the letter of 5 January 2018, i.e. around one month before the oral proceedings before the Board.

In said letter the amendments made to the respective claim 1 of these requests were identified. However no explanation was provided as to the purpose of the restrictions made, in particular how these would be directed to overcoming any of the objections with respect to inventive step raised by the appellant or to addressing the matters with respect to inventive step indicated by the Board in its communication. Even at the oral proceedings the submissions of the respondent regarding admittance of the requests were limited to stating that these requests constituted "fallback positions" with no further elucidation.

In view of the absence of any explanation as to the purpose of these amended requests, which it is emphasised, were provided after the summons to oral proceedings had been issued, the Board concluded that they were either irrelevant for the issue of inventive step and therefore their admittance did not have any impact on the outcome of the case, or that they raised new unknown issues, so that the Board and the appellant could not reasonably deal with these within the oral proceedings.

Consequently pursuant to Article 13(3) RPBA these requests are not admitted to the procedure.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. European patent No. 1754736 is revoked.

The Registrar:

The Chairman:



B. ter Heijden

D. Semino

Decision electronically authenticated