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Case Number: T 2060/16 - 3.3.10

Application Number: 11000010.6

Publication Number: 2308836

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Language of the proceedings: EN

Title of invention:
Polyisocyanate production method

Applicant:
Mitsui Chemicals, Inc.

Headword:
Polyisocyanate production method/ Mitsui Chemicals

Relevant legal provisions:
EPC Art. 56, 123(2)

Keyword:
Inventive step - (no)
Amendments - extension beyond the content of the application as filed (yes)

Decisions cited:

Catchword:

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Boards of Appeal

Chambres de recours

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Case Number: T 2060/16 - 3.3.10

D E C I S I O N
of Technical Board of Appeal 3.3.10
of 12 January 2021

Appellant: (Applicant)	Mitsui Chemicals, Inc. 5-2, Higashi-Shimbashi 1-chome Minato-ku Tokyo 105-7117 (JP)
Representative:	Mewburn Ellis LLP Aurora Building Counterslip Bristol BS1 6BX (GB)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted on 18 December 2015 refusing European patent application No. 11000010.6 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman	P. Gryczka
Members:	J.-C. Schmid
	T. Bokor

Summary of Facts and Submissions

I. The appeal lies from the decision of the Examining Division refusing European patent application 2308836.

II. According the examining division, claim 1 of the main request was modified by replacing "increasing an amount of" by "increasing the rate of production of". No basis for this amendment could be found in the application as filed. The term "increasing the rate" had not the same meaning as "increasing an amount". Consequently, this amendment present in claim 1 of the main request offended against Article 123(2) EPC.

As to inventive step, document (1) (US-A-2004/0024244) was the closest prior art to the invention. This document disclosed a process of producing polyisocyanates comprising a carbonyl chloride production process of producing carbonyl chloride by allowing chlorine to react with carbon monoxide, a polyisocyanate production process of producing polyisocyanate by allowing the carbonyl chloride produced in the carbonyl chloride production process to react with polyamine, and a chlorine production process of producing chlorine to be used in the carbonyl chloride production process by oxidizing hydrogen chloride produced secondarily in the polyisocyanate production process.

The technical problem was to set up a process to start such a reaction system. The problem was solved by starting each of the steps in a start-up operation and then performing a load-up operation in which the amount of production in one of the steps was increased and then the amount of production in the other two steps

was increased, this process being repeated until a desired steady state rate of production was achieved.

No reason could be seen why the skilled person would load up the reactions described in document (1) in any other way than was claimed. The start-up and load up procedures specified by claim 1 of the first auxiliary request were within the ambit of one of ordinary skill in the art and could not add anything inventive to the process disclosed in document (1).
The subject-matter of claim 1 of the first auxiliary request lacked therefore an inventive step.

Compared to the method of auxiliary request 1, the method of claim 1 of auxiliary request 2A incorporated a hydrochloric acid production process using both HCl produced secondarily in the isocyanate reactor and HCl not oxidized in chlorine production.

In the process disclosed in document (1), referring to figure 1, HCl produced secondarily in the isocyanate reactor **7** could be sent as stream **18** to a purification stage **19** involving absorption in water or dilute hydrochloric acid. Such a procedure inevitably brought about hydrochloric acid production from HCl produced secondarily in the isocyanate reactor and not sent to chlorine production. Furthermore a stream **23** exiting reactor **22** wherein chlorine was produced from HCl and oxygen contained HCl which was not oxidised in **22**. This stream was sent to apparatus **24** where dilute hydrochloric acid fed as **25** was converted to concentrated hydrochloric acid exiting **24** as **26** i.e. a net production of hydrochloric acid had occurred.
The added feature to claim 1 of the first auxiliary request covered this mode of operation, and thus did not add anything inventive to the subject-matter of

claim 1 of the first auxiliary request. Accordingly, the subject-matter of claim 1 of auxiliary request 2 did not meet the requirements of Article 56 EPC.

The examining division further rejected the then pending auxiliary requests 2, 3 and 3B for not complying with Article 123(2) EPC.

III. With the statement of grounds of appeal dated 28 April 2016, the Appellant filed a new main request and five auxiliary requests.

Claim 1 of the main request reads as follows:

"1. A polyisocyanate production method comprising:
a carbonyl chloride production process of producing carbonyl chloride by allowing chlorine to react with carbon monoxide,
a polyisocyanate production process of producing polyisocyanate by allowing the carbonyl chloride produced in the carbonyl chloride production process to react with polyamine, and
a chlorine production process of producing chlorine to be used in the carbonyl chloride production process by oxidizing hydrogen chloride produced secondarily in the polyisocyanate production process,

wherein the polyisocyanate production method comprises, in succession, (i) a start-up operation, (ii) a load-up operation in which the rate at which polyisocyanate is produced increases to a predetermined rate, and (iii) steady operation in which polyisocyanate is produced at said predetermined rate,

wherein the start-up operation is first performed by starting production of carbonyl chloride in the carbonyl chloride production process, starting production of polyisocyanate in the

polyisocyanate production process, and starting production of chlorine in the chlorine production process,

then the load-up operation, in which any one of (a) a process of increasing the rate of production of carbonyl chloride in the carbonyl chloride production process, (b) a process of increasing the rate of production of polyisocyanate in the polyisocyanate production process, and (c) a process of increasing the rate of production of chlorine in the chlorine production process, is selectively performed, and then the two other of said processes (a), (b) and (c) are performed, is repeatedly performed, until the rate of production of polyisocyanate reaches said predetermined rate."

Claim 1 of auxiliary request I differs from claim 1 of the main request by the polyisocyanate production method which comprises "(ii) a load-up operation in which the rate at which polyisocyanate is produced increases to a predetermined amount, and (iii) steady operation in which polyisocyanate is produced at said predetermined amount", and the load-up operation, *"in which any one of (a) a process of increasing an amount of production of carbonyl chloride in the carbonyl chloride production process, (b) a process of increasing an amount of production of polyisocyanate in the polyisocyanate production process, and (c) a process of increasing an amount of production of chlorine in the chlorine production process, is selectively performed, and then the two other of said processes (a), (b) and (c) are performed, is repeatedly performed, until an amount of polyisocyanate produced reaches said predetermined amount."*

Claim 1 of auxiliary requests II and III differs from claim 1 of the main request and auxiliary request I, respectively, in that the polyisocyanate production method comprises

"*a hydrogen chloride purification process of purifying hydrogen chloride produced secondarily in the polyisocyanate production process,*

a chlorine production process of producing chlorine to be used in the carbonyl chloride production process by oxidizing a large portion of hydrogen chloride supplied from the hydrogen chloride purification process, and

a hydrochloric acid production process of obtaining hydrochloric acid by absorbing, in water and hydrochloric acid water, unoxidized hydrogen chloride fed from the chlorine production process, and a part of hydrogen chloride fed from the hydrogen chloride purification process"

Claim 1 of auxiliary requests IV and V differs from claim 1 of auxiliary requests II and III, respectively, in that in the polyisocyanate production process, the process of increasing the rate of production of chlorine in the chlorine production process is operated "*by adjusting an amount of hydrogen chloride absorbed directly in water and hydrochloric acid water by regulating a volume of water supplied to the hydrochloric acid production process on the basis of the hydrogen chloride supplied from the chlorine production process and the hydrogen chloride supplied from the hydrogen chloride purification process or by regulating a quantity of hydrogen chloride supplied from the hydrogen chloride purification process on the basis of the hydrogen chloride supplied from the chlorine production process*".

IV. In a communication dated 24 September 2019, the Board indicated that claim 1 of the main and auxiliary requests 1 to 5 appeared not to meet the requirements of Article 123(2) or 76(1)) EPC.

V. In response to this communication, the Appellant further filed with letter dated 31 December 2019 auxiliary requests VI to VIII.

Claim 1 of auxiliary requests VI, VII and VIII differs from claim 1 of the main request and auxiliary requests II and IV, respectively, in that "*the polyisocyanate production method comprises, in succession, (i) a start-up operation, (ii) a load-up operation in which the amount of polyisocyanate being constantly produced increases to a predetermined amount, and (iii) steady operation in which polyisocyanate is constantly produced at said predetermined amount,*
and the load-up operation, in which any one of (a) a process of increasing an amount of production of carbonyl chloride in the carbonyl chloride production process, (b) a process of increasing an amount of production of polyisocyanate in the polyisocyanate production process, and (c) a process of increasing an amount of production of chlorine in the chlorine production process, is selectively *performed, and then the two other of said processes (a), (b) and (c) are performed, is repeatedly performed, until the amount of polyisocyanate being constantly produced reaches said predetermined amount.*"

VI. According to the Appellant, the application as filed described a continuous production process. In a continuous process, it was sensible to refer to the overall amount of the final product produced per unit time. As such, it was justifiable to use the phrase "rate of production" in the claims of the main request. This was how the skilled person would have anyway understood the invention. The use of the term "rate of production" was not intended to convey a different technical meaning over that of the application as filed. It was simply intended to more closely correspond to the invention as described in the

application as filed and as it would be understood by the skilled person. Accordingly, the use of the term “rate of production" in the claims of the main request and auxiliary requests did not constitute added subject-matter in comparison to the application as filed. The requirements of Article 123(2) EPC were met.

With regard to inventive step, document (1) represented the closest prior art to the invention. It disclosed a process which involved increasing the amount of polyisocyanate produced and the amount of chlorine produced after increasing the amount of produced carbonyl chloride, by increasing the supply of chlorine. This document did not describe a load-up operation for producing a polyisocyanate.

The process of the claim 1 of the main request therefore differed from that disclosed in document (1) in that a load-up operation was provided which involved selectively and sequentially executing, in the load-up operation, anyone of (a) increasing the rate of carbonyl chloride production in the carbonyl chloride production process, (b) increasing the rate of poly isocyanate production in the polyisocyanate production process and (c) increasing the rate of (recycled) chlorine production in the chlorine production process.

Since any one of the three processes could be selected as the one whose production amount was increased first in the load-up operation, and means for increasing the production amount in each process was operable, more effective operation of a polyisocyanate production system could be realized by selecting a production

amount increase in an appropriate process with appropriate timing.

Document (1) only suggested increasing a chlorine supply to increase the amount of carbonyl chloride produced and then increasing the amount of polyisocyanate produced and the amount of chlorine produced. Thus, in a start-up phase the chlorine used in the system was changed from externally added chlorine to recycled chlorine. A steady production state was established thereafter. This document neither disclosed nor suggested selectively first increasing a production rate in one process among the carbonyl chloride production, the polyisocyanate production and the recycled chlorine production in the load-up operation. The subject-matter of claim 1 of the main request involved therefore an inventive step.

Claim 1 of auxiliary requests II, III and VII required the presence of a hydrochloric acid production process which made use of hydrogen chloride from two sources: unoxidized hydrogen chloride from the chlorine production process, and hydrogen chloride derived directly from the polyisocyanate production process.

Thus, hydrogen chloride secondarily produced in the polyisocyanate production process was fed in parallel to a chlorine production process and a hydrochloric acid production process. The amount of hydrogen chloride gas supplied to each process could be easily regulated. This gave two advantages. First, regulating the supply of hydrogen chloride gas to the chlorine production process meant that the amount of chlorine produced in the chlorine production process could be easily adjusted. Second, regulating the supply of hydrogen chloride gas to the hydrochloric acid

production process meant that the concentration of hydrochloric acid produced in the hydrochloric acid production process could be easily adjusted.

Thus, the claimed method solved the additional technical problem of providing a hydrochloric acid stream at a concentration suitable for further industrial use.

Document (1) was not particularly concerned with the production of hydrochloric acid. Rather, document (1) was primarily concerned with a different technical problem, i.e. producing light-coloured isocyanates by reducing the amount of bromine and iodine-containing impurities incorporated into the isocyanates. This document failed to disclose or suggest that hydrogen chloride secondarily produced in a polyisocyanate production process was fed to both a chlorine production process and a hydrochloric acid production process in parallel.
Thus, the subject-matter of claim 1 of auxiliary request II, III and VII were not obvious in view of document (1).

In claim 1 of auxiliary requests IV, V, and VIII the hydrogen chloride secondarily produced in the polyisocyanate production process was fed to both the chlorine production process and the hydrochloric acid production process in parallel. Either the volume of water was regulated in the hydrochloric acid production process on the basis of unoxidized hydrogen chloride from the chlorine production process, and secondarily produced hydrogen chloride not fed to the chlorine production process; or the amount of the hydrogen chloride to be directly absorbed in the water and the hydrochloride acid water was adjusted by adjusting a

supply amount of secondarily produced hydrogen chloride not fed to the chlorine production process on the basis of the unoxidized hydrogen chloride from the chlorine production process.

Thereby, the concentration of hydrochloric acid could be regulated and further, a proportion of a supply of the hydrogen chloride secondarily produced in the polyisocyanate production process to each of the chlorine production process and the hydrochloric acid production process could be adjusted.

This enables adjustment of a hydrogen chloride supply to the chlorine production process, so that hydrogen chloride could be converted into chlorine at a constant conversion ratio in the chlorine production process, thereby increasing a chlorine production amount. Accordingly, the claimed method enabled a chlorine production amount to be increased with ease and enabled effective operation of a polyisocyanate production system in the load up operation.

In document (1), hydrogen chloride could be supplied directly to each of the hydrogen chloride oxidation reactor and the phase contact apparatus from the hydrogen chloride purification process. Hydrogen chloride was supplied to the phase contact apparatus **24** only after the hydrogen chloride had passed through the hydrogen chloride oxidation reactor **22**.

Accordingly, the rate of production of chlorine could not be adjusted by regulating the amount of hydrogen chloride supplied to each of the chlorine production process and the hydrochloric acid production process.

The subject-matter of claim 1 of auxiliary requests IV, V and VIII involved therefore an inventive step.

VII. The Appellant (applicant) requested that the decision under appeal be set aside, and a patent be granted on the basis of claims 1-7 of the Main request, or alternatively on the basis of one of the Auxiliary Requests I to V, filed with the grounds of appeal dated 28 April 2016, or on the basis of one of the Auxiliary Requests VI to VIII, filed with letter dated 31 December 2019.

VIII. At the end of the oral proceedings held on 12 January 2021, the decision of the Board was announced.

Reasons for the Decision

1. The appeal is admissible.

Main request and auxiliary requests II and IV

2. *Article 123(2) EPC*

With respect to claim 1 as originally filed, the load up operation in claim 1 of these requests has been modified *inter alia* to comprise "(a) a process of increasing the **rate** of production of carbonyl chloride … until the **rate** of production of polyisocyanate produced reaches said predetermined **rate**", whereas the load up operation of originally filed claim 1 comprised "a process of increasing an amount of carbonyl chloride produced … until an amount of polyisocyanate produced reaches a predetermined amount".

There is no basis for this amendment in the application as filed, since the term "rate" does not appear in the application as filed.

According to the applicant, where a process is implemented on a continuous basis, it was more sensible to refer to the overall quantity of the product manufactured per unit of time, rather than to an amount of production. Consequently, the skilled person would understand that "an amount of production" means "the rate of production" in the context of the invention.

However, what the skilled person finds evident from a disclosure and the content of the disclosure itself are two distinct considerations. Accordingly, even assuming that it would be obvious for the skilled person that load up operation should be understood to comprise increasing the rate of production of carbonyl chloride … until the rate of production of polyisocyanate produced reaches said predetermined rate, this is not disclosed in the application as filed.

Moreover, if these two expressions had the same meaning in the context of the invention, it would not be necessary to change "an amount of production" to "the rate of production" in claim 1. Accordingly, the main request and auxiliary requests II and IV must be rejected for not complying with the requirements of Article 123(2) EPC.

Auxiliary requests I, III and V

3. Claim 1 of these requests requires a load-up operation in which the **rate** at which polyisocyanate is produced increases to a predeterminate amount, for which no basis can be found in the application as filed.

Accordingly, these requests must be rejected too for not complying with the requirements of Article 123(2) EPC.

Auxiliary request VI

4. *Amendments*

Claim 1 of this request corresponds to claim 1 of the main request but returns to the original language referring to an amount of production rather than a rate of production.
The Board is therefore satisfied that claim 1 of auxiliary request VI satisfies the requirements of Article 123(2) EPC.

5. *Inventive step*

Document (1) relates to a continuous process of preparing organic isocyanate comprising the step of
(c) producing carbonyl chloride by reacting chlorine with carbon monoxide,
(d) reacting carbonyl chloride with a primary amine to produce the corresponding isocyanate and hydrogen chloride; and,
(f) purifying the hydrogen chloride formed in step (d), and
(g) producing chlorine by oxidizing hydrogen chloride.

Document (1) discloses a process for the preparation of polyisocyanates (claim 9) in which chlorine and CO are reacted to give carbonyl chloride (claim 1; step (c)), the obtained carbonyl chloride is reacted with a polyamine to give the desired polyisocyanate and hydrogen chloride as co-product (step (d)), which is

purified (step (f)) to produce chlorine by oxidation with oxygen (step (g)). The chlorine thus obtained is then combined with an external source of chorine to produce carbonyl chloride (steps (h) and (c)). The process of document (1) is therefore a loop in that a product of each stage is used in the next stage, with a product of the last stage being used in the first stage (also see figure 1). The process may be operated under steady-state conditions (see paragraph [0028].

Thus, document (1) discloses the polyisocyanate production method of claim 1 of auxiliary request VI, except those operations that must be implemented before reaching steady state conditions. Hence, in order to carry out the continuous process disclosed in document (1), the skilled person must fill the gap concerning the start of the process with his general knowledge.

Paragraph [0027] of document (1) discloses that at start-up of the procedure the first and second partial amounts of chlorine mentioned come from the same source. It cannot be from the oxidation of HCl in the third step of the process as this situation only begins after the start-up phase. This leaves only the external source of chlorine as the mode of starting the reaction. The skilled person would thus logically start from the production of carbonyl chloride and then would start the other steps sequentially when the respective starting products become available.

Document (1) also indicates that during the start up the carbonyl chloride is prepared by using the chlorine only from an external source (page 2, right-hand column, lines 2 to 4) and then, when the process operation has passed the start-up phase and sufficient chlorine is available from step (h), the chlorine used

to prepare carbonyl chloride will have a significantly lower bromine and iodine content (see page 2, right-hand column, lines 4 to 12).

This indicates that the production of chlorine in step (h) with low degree of impurities increases gradually, with the consequence that there is also a loading up for the other steps which are gradual too. Moreover gradually increasing the load is common practice for the implementation of continuous processes on a large scale, as the control of a reaction starting with a full load is impractical.

The skilled person would logically operate the load-up sequentially by increasing the amount of starting material when more material is formed from the previous step. The skilled person would thus arrive at the subject-matter of claim 1 of auxiliary request 6 without involving an inventive step.

According to the Appellant, the claimed method provides flexibility, since any of the steps (a), (b) or (c) could selectively be chosen to increase the amounts.

However, this is more a question of the scope of the claim in terms of the number of alternatives claimed, rather than a question of flexibility. In fact, claim 1 seems to cover almost all practical alternatives for implementing the starting/loading operations of the process of document (1). In any event, claim 1 includes at least the obvious alternative of carrying out the sequential starting/loading operation as described above.

Hence, the argument relating to the improvement of flexibility does not convince the Board.

Auxiliary request VI is thus rejected for lack of inventive step.

Auxiliary Request VII

6. The process of claim 1 of auxiliary request VII additionally comprises a purification step of the hydrogen chloride produced secondarily in the polyisocyanate production process, and a hydrochloric acid production process by absorbing, in water and hydrochloric acid water, unoxidized hydrogen chloride fed from the chlorine production process, and a part of hydrogen chloride fed from the hydrogen chloride purification process

The feature of purifying the hydrogen chloride obtained in the polyisocyanate production process does not add anything new with respect to the process of document (1) wherein the hydrochloric acid produced secondarily in the polyisocyanate production process of document (1) is also purified (see figure 1, purification stage **19**).

In the process of document (1) the unoxidized hydrogen chloride fed from the chlorine production process **23** is brought into contact with dilute hydrochloric acid **25** to produce a more concentrated hydrochloric acid **26.**

Hence, apart from the start-up and load-up procedures, the process of claim 1 of auxiliary request VII further differs from the process disclosed in document (1) only in that a portion of the purified hydrogen chloride produced secondarily in the polyisocyanate production is used to produce concentrated hydrochloric acid.

Document (1) is concerned with the production of light-coloured isocyanates. The colour of isocyanates is mainly caused by the presence of bromide and iodide impurities in the chlorine used to prepare carbonyl chloride (see paragraphs [0001] to [0003]). The purification required to reduce the bromine and iodine content of the chlorine is associated with high costs (see paragraph [0005]). Document (1) aims to reduce the purification of the raw material used in the production of isocyanate (see paragraph [0007]). Hence, since hydrogen chloride produced as a by-product in the production of isocyanates is practically free of bromine or iodine, document (1) proposes to use this chlorine (second source), to be mixed with external chlorine having much higher bromine and iodine content (first source), in the production of chlorine with an acceptable content of impurities (see paragraph [0027]), thereby reducing the need for full purification of the external chlorine supplied to the process.

However, if the skilled person does not wish to avoid complete purification of the external chlorine introduced into the process, it will recognise that the purified hydrogen chloride produced as a by-product in the production of polyisocyanate does not need to be wholly recycled into the reaction, but at least a part of it, can be used for other purposes, for example in the preparation of industrial concentrated hydrochloric acid.

No inventive step can be acknowledged for using purified HCl produced as a by-product during the polyisocyanate production in the preparation of concentrated hydrochloric acid rather than in the preparation of Cl_2.

The subject-matter of claim 1 of auxiliary request VII lacks therefore an inventive step.

According to the Appellant, splitting the stream of the purified hydrogen chloride produced secondarily in the polyisocyanate production provides advantages in that the process could be better controlled, e.g. the amount of recycled chlorine could be simply and easily adjusted by adjusting the amount of HCl fed in hydrochloric acid production.

However, the skilled person will easily recognize that these advantages are associated with a fractionation of the purified hydrogen chloride produced secondarily in the polyisocyanate production. Therefore, this argument does not convince the Board.

Consequently, auxiliary request VII is rejected for lack of inventive step.

Auxiliary request VIII

7. In the process of claim 1 of auxiliary request VIII, the process of increasing the amount of production of chlorine in the chlorine production process is limited to two alternatives.

The second alternative of the claim consists in regulating a quantity of hydrogen chloride supplied from the hydrogen chloride purification process on the basis of the hydrogen chloride supplied from the chlorine production process.

In the process of document (1), chlorine is obtained by oxidizing HCl coming from two sources, the first coming from the purification stage (stream **20**) and the other

from the chlorine production process (stream **37**; recycled HCl).

The means of increasing the chlorine production by regulating the quantity of hydrogen chloride supplied from the hydrogen chloride purification process on the basis of the hydrogen chloride supplied from the chlorine production process is an obvious option for the skilled person.

As the second alternative is obvious in the light of document (1), it is not necessary to consider the other claimed alternative.

Consequently, auxiliary request VIII must also be rejected for lack of inventive step.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

C. Rodríguez Rodríguez P. Gryczka

Decision electronically authenticated