

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

PCT

**WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY
(PCT Rule 43bis.1)**

To:

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Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference
see form PCT/ISA/220

FOR FURTHER ACTION
See paragraph 2 below

International application No.
PCT/EP2018/073601

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International Patent Classification (IPC) or both national classification and IPC
INV. C01B39/30 B01D53/04 B01J29/50 B01J29/56 C01B39/02 C07C1/20

Applicant
HALDOR TOPSOE AS

1. This opinion contains indications relating to the following items:

- Box No. I Basis of the opinion
- Box No. II Priority
- Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- Box No. IV Lack of unity of invention
- Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- Box No. VI Certain documents cited
- Box No. VII Certain defects in the international application
- Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Name and mailing address of the ISA:



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Date of completion of this opinion

see form
PCT/ISA/210

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Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:
 - the international application in the language in which it was filed.
 - a translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1 (b)).
2. This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(a))
3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing:
 - a. forming part of the international application as filed:
 - in the form of an Annex C/ST.25 text file.
 - on paper or in the form of an image file.
 - b. furnished together with the international application under PCT Rule 13ter.1(a) for the purposes of international search only in the form of an Annex C/ST.25 text file.
 - c. furnished subsequent to the international filing date for the purposes of international search only:
 - in the form of an Annex C/ST.25 text file (Rule 13ter.1(a)).
 - on paper or in the form of an image file (Rule 13ter.1(b) and Administrative Instructions, Section 713).
4. In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that forming part of the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
5. Additional comments:

Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	<u>10, 11, 13-20, 23, 24</u>
	No: Claims	<u>1-9, 12, 21, 22</u>
Inventive step (IS)	Yes: Claims	
	No: Claims	<u>1-24</u>
Industrial applicability (IA)	Yes: Claims	<u>1-24</u>
	No: Claims	

2. Citations and explanations

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

1 Reference is made to the following documents:

- D1 JOO HYUCK LEE ET AL: "Synthesis and Characterization of ERI-Type UZM-12 Zeolites and their Methanol-to-Olefin Performance", JOURNAL OF THE AMERICAN CHEMICAL SOCIETY, AMERICAN CHEMICAL SOCIETY, US, vol. 132, no. 7, 22 September 2010, pages 12971-12982, XP002668581
& JOO HYUCK LEE ET AL: "Supporting info for Synthesis and Characterization of ERI-Type UZM-12 Zeolites and their Methanol-to-Olefin Performance", XP055513517
- D2 JIE ZHU ET AL: "Ultrafast synthesis of high-silica erionite zeolites with improved hydrothermal stability", CHEMICAL COMMUNICATIONS, vol. 53, no. 50, 24 May 2017, pages 6796-6799, XP055513320,
& Jie Zhu ET AL: "Supporting information for Ultrafast synthesis of high-silica erionite zeolites with improved hydrothermal stability", Chemical communications (Cambridge, England), XP055513700
- D3 MARTÍN NURIA ET AL: "Cage-based small-pore catalysts for NH₃-SCR prepared by combining bulky organic structure directing agents with modified zeolites as reagents", APPLIED CATALYSIS B: ENVIRONMENTAL, ELSEVIER, AMSTERDAM, NL, vol. 217, 29 May 2017, pages 125-136, XP085112832
- D4 US 2006/073094 A1 (MILLER MARK A [US] ET AL) 6 April 2006
- D5 US 2016/001273 A1 (XIE DAN [US] ET AL) 7 January 2016 (2016-01-07) - cited in the application

2 Re Item VIII

Certain observations on the international application

2.1 **Clarity (Art. 6 PCT)**

The application does not meet the requirements of Article 6 PCT, because **claims 1-24** are not clear.

- 2.1.1 It is not clear from how **claim 1** is currently written if a calcined ERI or an as-synthesized ERI is intended. As it is known in the field of catalysis that the calcined form should be used as otherwise the pores are blocked by the template, for this WO-ISA, the assumption will be made that the template has been removed from the material, and it is thus in its calcined form.

The same is valid for all dependent **claims 2-6**.

- 2.1.2 Present **claim 1** relates to a product defined (inter alia) by reference to the following unusual parameter:

a crystal morphology defined by the ratio between the dimensions r_c along and r_a orthogonal to the unique c-axis between 0.5 and 2.0

The use of this unusual parameter in the present context is considered to lead to a lack of clarity because the claim does not clearly identify the products encompassed by it as, although the parameter can be clearly and reliably determined by indications in the description, this procedure is not at all usual in the art which makes it impossible to compare the claim to the prior art. As a result, the application does not comply with the requirement of clarity under Article 6 PCT.

The same is valid for all dependent **claims 2-6**.

- 2.1.3 **Claim 1** is not supported by the description as required by Article 6 PCT, as its scope is broader than justified by the description.

The present claim 1 relates to an extremely large number of possible materials as *any* catalyst that comprises an ERI zeolite is envisaged as long as it has a molar ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ of 8-100 and *a crystal morphology defined by the ratio between the dimensions r_c along and r_a orthogonal to the unique c-axis between 0.5 and 2.0*, which is an unusual parameter (see point 2.1.2)

Support and disclosure within the meaning of Articles 5 and 6 PCT are to be found, however, for only a very small proportion of the materials claimed i.e. a catalyst comprising an ERI-type structure having a molar ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ up to 22.0 as shown in all examples 2-5 of the description.

It has only been demonstrated that an ERI material can be made having a molar ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ up to 22.0. The synthesis of ERI zeolitic materials is a very unpredictable science, i.e. what works for one type of material might not work for another type, it is highly unlikely that the same process will work and lead to merely any molar ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ between 8-100.

Claim 1 is not regarded as supported by the description as the skilled person would not be able, on the basis of the information as filed, to extend the particular teaching of the examples to the whole of the content of claim 1, by using routine experimentation (Article 6 PCT).

The same is valid for **claims 2-6** as they are dependent on claim 1 and for **claims 7-24**, as they are related to claim 1.

This problem can also be considered as an objection of lack of disclosure of the invention under Article 5 PCT, in the sense that only specific ways of carrying out the invention are exemplified. The information given in the description is of such limited nature that the skilled person doesn't have sufficient clear information to reduce in practice without undue burden, with a reasonable expectation of success, which features/solutions solve the problem and which do not.

2.1.4 The term '*about*' used in **claim 1** is vague and unclear and leaves the reader in doubt as to the meaning of the technical feature to which it refers, thereby rendering the definition of the subject-matter of said claim unclear (Article 6 PCT).

2.2 **Miscellaneous**

2.2.1 **Claim 4** includes all the features of claim 3. Hence, claim 4 should be reformulated as a claim dependent on claim 3 (Rule 6.4 PCT).

2.2.2 There appears to be a typographic error in **claim 21** line 28, as the word *method* seems to be missing after *the*.

3 **Re Item V**

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

3.1 **Novelty (Art. 33(2) PCT)**

The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of **claims 1-9, 12, 21 and 22** is not new in the sense of Article 33(2) PCT.

3.1.1 Concerning **claim 1** document D1 discloses:

- a catalyst comprising a molecular sieve with the ERI framework type (UZM-12 in D1, Tables 1 and 3 and Fig.1, p.12973 right-hand column third paragraph)
- having a mole ratio of silica to alumina from about 8 to about 100 (D1, Table 3 provides Si/Al ratios between 4.7 and 6.5 which corresponds with silica/alumina ratios between 9.4 and 13) and

- a crystal morphology defined by the ratio between the dimensions r_c along and r_a orthogonal to the unique c-axis between 0.5 and 2.0 (as this is an unusual parameter, this value is not retrieved in the prior art (see point 2.1.2), however, the SEM images provided in D1 show morphologies which are similar to those shown in the present application, therefore this technical feature is also anticipated by D1 (D1, fig.3, Table 5 and supporting info fig. S1)

Therefore, the subject-matter of **claim 1** lacks novelty (Art. 33(2) PCT).

3.1.2 The additional features set out in dependent **claims 2-4** are also anticipated by document D1:

- **claim 2:** the calcined form of the molecular sieve has a powder X-ray diffraction pattern collected in Bragg-Brentano geometry with a variable divergence slit using Cu K-alpha radiation essentially as shown in the Table (calcined UZM-12 that is disclosed in D1, Tables 1 and 3 and Fig. 1 and p.12972 right-hand column last paragraph)
- **claim 3:** wherein the silica-to-alumina mole ratio is between 8 and 100 (D1, Table 3 provides Si/Al ratios between 4.7 and 6.5 which corresponds with silica/alumina ratios between 9.4 and 13)
- **claim 4:** wherein the silica-to-alumina mole ratio is between 10 and 60 (D1, Table 3 provides Si/Al ratios between 4.7 and 6.5 which corresponds with silica/alumina ratios between 9.4 and 13).

Therefore, the subject-matter of **claims 2-4** lacks novelty (Art. 33(2) PCT).

3.1.3 Concerning **claim 21** document D1 discloses:

- a method for the conversion of oxygenates to hydrocarbons the comprising the step of contacting the oxygenates with a catalyst according to any one of claims 1 to 6 (D1, p.12973 right-hand column third paragraph, p.12979 right-hand column last paragraph - p.12980 first paragraph, fig.7-10)

Therefore, the subject-matter of **claim 21** lacks novelty (Art. 33(2) PCT).

3.1.4 The additional features set out in dependent **claim 22** are also anticipated by document D1:

- **claim 22:** the produced hydrocarbons comprise olefins (D1, p.12973 right-hand column third paragraph, p.12979 right-hand column last paragraph - p.12980 first paragraph, fig.7-10)

Therefore, the subject-matter of **claim 22** lacks novelty (Art. 33(2) PCT).

3.1.5 For sake of completeness, it is pointed out that the subject-matter of **claims 1-4** is also anticipated by documents D2, D3 and D4

(see the respective passages cited in the international search report for each of these documents).

3.1.6 The additional features set out in dependent **claims 5 and 6** are also anticipated by document D3 and/or D5:

- **claims 5:** wherein at least a part of the aluminum and/or silicon is substituted by one or more metals selected from tin, zirconium, titanium, hafnium, germanium, boron, iron, indium and gallium (D5, par.[0006] and [0017]).
- **claim 6:** containing copper and/or iron (D3, p.127-128 points 2.2.1 and 2.2.2 and Table 3; D5, par.[0006] and [0017]).

Therefore, the subject-matter of **claims 5 and 6** lacks novelty (Art. 33(2) PCT).

3.1.7 Concerning **claims 7 and 12** document D3 discloses:

- **claim 7:** a method for the conversion of nitrogen oxides to nitrogen in presence of a reductant comprising the step of contacting the nitrogen oxides and the reductant with the catalyst according to any one of claims 1 to 6 (D3, p.129 left-hand column point 2.5 Catalytic testing and p.131 left-hand column last paragraph to p.135 left hand column second paragraph, Catalytic performance in NH₃-SCR of NO, fig.9 and 14).
- **claim 12:** a method for the selective oxidation of ammonia to nitrogen comprising the step of contacting the ammonia or a gas comprising the ammonia with the catalyst according to any one of claims 1 to 6 (D3, p. 129 left-hand column point 2.5 Catalytic testing and p.131 left-hand column last paragraph to p.135 left hand column second paragraph, Catalytic performance in NH₃-SCR of NO, fig.9 and 14).

Therefore, the subject-matter of **claims 7 and 12** lacks novelty (Art. 33(2) PCT).

3.1.8 The additional features set out in dependent **claim 8** are also anticipated by document D3:

- **claim 8:** the reductant comprises hydrocarbons and/or ammonia or a precursor thereof (ammonia in D3, p.129 left-hand column point 2.5 Catalytic testing and p.131 left-hand column last paragraph to p.135 left hand column second paragraph, Catalytic performance in NH₃-SCR of NO, fig.9 and 14).

Therefore, the subject-matter of **claim 8** lacks novelty (Art. 33(2) PCT).

- 3.1.9 For sake of completeness, it is pointed out that the subject-matter of **claims 7, 21 and 22** is also anticipated by document D4

(see the respective passages cited in the international search report for this document).

- 3.1.10 The additional features set out in dependent **claim 9** are also anticipated by document D4:

- **claims 9:** the nitrogen oxides are contained in engine exhaust (D4. par. [0049])

Therefore, the subject-matter of **claim 9** lacks novelty (Art. 33(2) PCT).

3.2 ***Inventive step (Art. 33(3) PCT)***

The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of **claims 10, 11, 13-20, 23 and 24** does not involve an inventive step in the sense of Article 33(3) PCT.

- 3.2.1 Use **claims 18, 23 and 24** relate to the use of the ERI catalyst. As these uses of ERI zeolites are well-known (see for example D5, par.[0059], [0064]-[0067], [0073]). The use of the known ERI material of the current application (see points 3.1.1-3.1.10) for the same purpose therefore cannot be considered to involve an inventive step. Therefore, the subject-matter of **claims 18, 23 and 24** is not inventive (Article 33(3) PCT).

- 3.2.2 Dependent **claims 10, 11, 13-17 and 19-20** do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of inventive step in that the additional features of **claims 10, 11, 13-17 and 19-20** are already known and/or appear to represent mere design options obvious to the person skilled in the art which are not associated with an unexpected technical effect that could support the presence of an inventive step (see passages cited in the search report for documents D1-D5).