

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:

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PCT

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

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/US2022/076047

International filing date (day/month/year)
07.09.2022

Priority date (day/month/year)
09.09.2021

International Patent Classification (IPC) or both national classification and IPC
INV. C10L3/12 C07C1/02 C07C1/04 C07C1/12 C07C9/08 C07C9/10 C01B3/38 C10K3/02

Applicant
GAS TECHNOLOGY INSTITUTE

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.

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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.
 - b. ☐ furnished subsequent to the international filing date for the purposes of international search (Rule 13ter.1(a)),
 - ☐ accompanied by a statement to the effect that the sequence listing does not go beyond the disclosure in the international application as filed.
4. ☐ With regard to any nucleotide and/or amino acid sequence disclosed in the international application, this opinion has been established to the extent that a meaningful opinion could be formed without a WIPO Standard ST.26 compliant sequence listing.
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>2-4, 19, 20, 22</u>
	No: Claims	<u>1, 5-18, 21</u>
Inventive step (IS)	Yes: Claims	
	No: Claims	<u>1-22</u>
Industrial applicability (IA)	Yes: Claims	<u>1-22</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

Re Item V

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

- 1 Reference is made to the following documents:
D1: WO 2004/076600 A
D2: WO 2005/037962 A
D3: CN 110 846 096 A
D4: CN 101 658 805 A
D5: EP 0 146 165 A
- 2 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1, 5-18, 21 is not new in the sense of Article 33(2) PCT.
 - 2.1 Document D1 (see the passages cited in the International search report, particularly figure 1 and example 1, reference is made to the attached machine translation) discloses a process for producing LPG product comprising propane and butane in a two stage reaction comprising (a) a reforming reaction to generate synthesis gas and (b) an LPG production reaction. Effluent from the LPG production is split into an LPG product stream and a stream recycled to the inlet of the reformer. In example 1, a gas composition comprising 7.8 mol% of natural gas, 14.3 mol% of steam, 3.9 mol% of carbon dioxide and 74 mol% of a recycle stream is fed to the reformer reactor. The recycle stream comprises 0.6 mol% methane, 0.8 mol% ethylene, 39 mol% carbon dioxide, 22 mol% carbon monoxide and 38 mol% hydrogen. From this, the combined stream is calculated to comprise 32,76 mol% CO₂, 8,244 mol% CH₄, 28,12 mol% H₂, 16,28 mol% CO and 4,3% H₂O (natural gas was counted as methane, numbers are not rounded). The stream thus comprises predominantly H₂ and CO₂ and (60.88 mol%). The intermediate syngas comprises 44 mol% of hydrogen, 43 mol% of carbon monoxide, 6 mol% of carbon dioxide and 7 mol% of methane. The effluent of the second stage is dried and separated into the LPG product and the recycle stream. It is further stated that 70% of the hydrocarbons produced in the second stage were propane and butane, and that the propane and butane content was 41% propane and 59% butane, based on carbon. In the first stage, a Ru/MgO catalyst is used, which is evidently capable of catalysing both reforming and RWGS reactions (CO₂ is consumed in the presence of H₂). In the second stage, comprising a mixture of a commercial Cu-Zn methanol synthesis catalyst and zeolite H-ZSM-5 (a dehydration catalyst in the sense of the

- application, see para. [17]) is used as the catalyst system. Document D1 discloses the subject-matter of independent process claims 1, 16, 21. It further discloses the additional technical features of dependent claims 5-12, 17, 18. Therefore, the subject-matter of claims 1, 5-12, 16-18, 21 is not novel over document D1.
- 2.2 Document D2 (see the passages cited in the International search report, particularly figure 2 and example 2, reference is made to the attached machine translation) discloses a process for producing LPG product comprising propane and butane in a two stage reaction comprising (a) a reforming reaction to generate synthesis gas and (b) an LPG production reaction. In one embodiment, effluent from the LPG production is split into an LPG product stream and a stream recycled to the inlet of the LPG reactor. In example 2, a gaseous feed mixture comprising 45 mol% of natural gas, 45 mol% of steam and 10 mol% of carbon dioxide is fed to the reformer reactor. The gaseous feed mixture thus comprises predominantly CH₄ and CO₂ (55 mol%, counting natural gas as methane). The syngas produced in the reformer comprises 62 mol% of hydrogen, 31 mol% of carbon monoxide, 4.8 mol% of carbon dioxide and 2.2 mol% of methane. A combined stream of syngas and recycle separated from the LPG reactor effluent is fed into the LPG reactor, the combined stream comprising 51 mol% hydrogen, 26.9 mol% carbon monoxide, 14.2 mol% carbon dioxide, 6.9 mol% methane and 1.0 mol% others (see original para. [0195], the machine translation fails to properly indicate 51 mol% hydrogen). The effluent of the second stage is dried and separated into the LPG product and the recycle stream. It is further stated that 75% of the hydrocarbons produced in the second stage were propane and butane (split: 56% propane and 44% butane, see original para. [0196]). The recycle stream contains, *inter alia*, 25.9 mol% of carbon dioxide and 37.7 mol% of hydrogen (original para. [0197]). The reforming stage employs a Ru/MgO catalyst, while the second stage uses a mixture of a commercial Cu-Zn methanol synthesis catalyst and zeolite H-ZSM-5. Document D2 is novelty-destroying in respect of claims 1, 5, 6, 8-12, 16-18, 21.
- 2.3 Independent claim 13 is directed to an LPG product defined by its renewable carbon content. Claim 13 is unrelated to the process of claim 1. Furthermore, claim 1 does not contain any renewable carbon content limitation. The only common matter of claims 1 and 13, an LPG product, is not only specifically disclosed in documents D1 and D2, but also part of the skilled person's common general knowledge. Furthermore, the remaining features (LPG production process in claim 1, renewable carbon content in claim 13) do not establish a

single general inventive concept. It is therefore *prima facie* evident that claims 1 and 13 lack unity of invention. In this instance, however, this Authority considers that a full objection under Rule 13 PCT is not warranted because claims 13-15 manifestly lack novelty and an inventive step under Article 33 PCT. Document D3 (see the cited passages, particularly claim 1, reference is made to the attached machine translation) relates to LPG obtained from biomass synthesis gas. The biomass synthesis gas is obtained by the gasification of biomass (para. [0003]-[0005]) which, in the absence of any indication to the contrary, must be understood as containing only renewable carbon. This property is preserved in the LPG product exclusively produced from the carbon contained in the biomass synthesis gas. Consequently, D3 discloses an LPG product within the scope of claim 13. In view of the non-limiting character of the additional technical features of dependent claims 14, 15 (see Item VIII below), these claims are not novel either.

- 3 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-22 does not involve an inventive step in the sense of Article 33(3) PCT. The additional technical features of dependent claims 2-4, 19, 20, 22, for which novelty is acknowledged, do not establish any inventive difference over the prior for the following reasons.
- 3.1 Regarding claim 2, no surprising or unexpected technical effect has been shown for a higher amount of any one of (i) CH₄ and CO₂ or (ii) H₂ and CO₂ in the gaseous feed mixture. It is noted that FEED A and FEED C used in the examples (incidentally having a combined H₂ and CO₂ content above 75 mol-%) are synthesis gas intermediates to be converted in the LPG synthesis stage (b) within the meaning of claim 1, while claim 2 defines the gaseous feed mixture to be converted in the reforming or RWGS stage (a). It is considered that a higher content of H₂ and CO₂, which are the principal constituents of the recycle stream of D1, would be achieved within the overall teaching of D1 by routine variation of conversion and recycle rates. Finally, it is noted that D2 discloses a reaction sequence in which the production of syngas and the synthesis of LPG are sequential and unrelated (unlike in D1, the recycle of D2 does not have an influence on the synthesis gas production stage). They are thus a mere juxtaposition of independent processes, just like the in present application (see para. [18]). The skilled person would therefore consider alternative synthesis gas production routes as the circumstances require. Synthesis gas processes involving dry reforming or RWGS are known from the prior art (see the cited

- passages of document D4 for dry reforming of biogas, notably para. [0119], reference is made to the attached machine translation) and would be considered as obvious alternatives. No inventive step is involved.
- 3.2 The juxtaposition argument and the alternative syngas production proposed in D4 also render the subject-matter of claims 3, 4 obvious and non-inventive.
- 3.3 Biogas defined in claims 4, 19 is an obvious alternative in the case of both D1 (example feed containing 7.8% CH₄ and 3.9% CO₂) and D2 (example feed containing 45% methane and 10% CO₂), knowing that biogas contains a significant amount of CO₂ in addition to CH₄ (D4 para. [0119]). It is noted that the use of biogas inevitably provides the LPG product of claims 13-15, which for this reason lack an inventive step, too.
- 3.4 Claim 20 is not inventive because electrical reactor heating is commonplace and would be provided by the skilled person according to the needs.
- 3.5 Claim 22 defines yttrium-containing catalysts in the LPG synthesis stage starting from synthesis gas. Only para. [75] contains speculative statements on alleged technical effects of yttrium, irrespective of the catalyst to which it is added, which are not supported by evidence. Methanol synthesis catalysts comprising copper and zinc as well as yttrium as a rare earth metal modifier are as such known from document D5 (claims 1, 5, 6, 12) and would be employed in the processes of D1 or D2 (both utilising a copper-zinc methanol synthesis catalyst in the LPG synthesis step) as an obvious and non-inventive alternative.

Re Item VIII

Certain observations on the international application

- 4 The application does not meet the requirements of Article 6 PCT, because claims 1, 8, 9, 12, 14, 15 are not clear.
- 4.1 Claim 1 lacks clarity because the term "predominantly" is undefined. The definition of para. [34] or claim 2 should be inserted.
- 4.2 Claim 8 is unclear as the term "methanol synthesis mechanism" is undefined. It is not clear whether methanol should be an identifiable intermediate or even present in the final product, or whether it suffices that the same type of catalysts also effective in methanol synthesis are employed.

- 4.3 In claim 9, the difference between alternatives (i) and (ii) is not clear. It is noted that a catalyst mixture also constitutes a bi-functional catalyst in the claimed sense.
- 4.4 Claim 12 is unclear because the basis on which the feed carbon content should be calculated is not unambiguously defined. The gaseous feed mixture may contain other carbon sources than CH₄ and CO₂, most notably CO (see claim 3). It may also contain other hydrocarbons, especially in situations where it comprises a recycle portion (see claim 7). It is not defined whether these other carbon sources should be included in the calculation of the 70% figure.
- 4.5 Claims 14, 15 contain product-by-process type definitions. In this instance, the intended limitation cannot be clearly established. The claims relate to an LPG product *per se*, irrespective of how it is produced. However, the technical features of claims 14, 15 pertain to the starting materials. These have no structural impact on the LPG product itself.