Case Number: T 0294/95 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 31 January 1996

Appellant: THE BOC GROUP, INC.
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 6 December 1994 refusing European patent application No. 91 311 141.5 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: M. Chomentowski
B. J. Schachenmann

Summary of Facts and Submissions

I. European patent application No. 91 311 141.5 (publication No. 0 489 555) was refused on the grounds of lack of inventive step of the subject-matter of the independent claims having regard to D1 = EP-A-0 317 235 and D2 = US-A-4 917 711.

The Examining division took the following view:

In the pressure swing adsorption (PSA) process of D1 for producing carbon monoxide and hydrogen from a feed mixture comprising additionally carbon dioxide and methane, the adsorbents used show a higher affinity for carbon dioxide than for carbon monoxide; since the carbon monoxide is produced in a second PSA step in a second PSA plant, the method is complicated. There is no doubt that a person skilled in the art of adsorption will try to find a simpler one and it is part of his basic knowledge that a desired component of a gas stream can be separated by a (simpler) one step process when this product shows the highest or the lowest adsorption affinity of all gases in the gas stream for a given adsorbent. As D2 mentions an adsorbent showing a higher affinity for carbon monoxide than for all other components and in particular carbon dioxide, the skilled person will apply such an adsorbent, whereby one of the PSA steps of D1 becomes superfluous. Therefore, since further features distinguishing the claimed processes are only features known from D1 and/or features which are to be used for the system to function, said claimed processes lack an inventive step.

II. The appellant (applicant) lodged an appeal against this decision.

III. During the oral proceedings of 31 January 1996 which had been requested auxiliarily, the appellant filed a new set of 5 claims and requested that the decision under appeal be set aside and a patent be granted on the basis of said claims, with the description and drawings to be adapted. The only independent claim reads as follows:

"1. A method of producing by pressure swing adsorption hydrogen and carbon monoxide products from a feed mixture comprising hydrogen, carbon monoxide, carbon dioxide, and methane, characterised by repeating the steps of:

(a) providing a pressure swing adsorption system (M) having a first stage and a second stage, wherein the first stage contains an adsorption bed comprising an adsorbent having a greater affinity for carbon monoxide than for hydrogen, carbon dioxide, and methane, the second stage contains an adsorption bed comprising an adsorbent having a greater affinity for carbon dioxide, methane, and carbon monoxide than for hydrogen, and the first and second stages are connected in series and each stage contains an inlet end and an outlet end;

(b) passing the feed mixture through the inlet end of the first stage of the pressure swing adsorption system (M) to separate carbon monoxide as an adsorbed fraction and hydrogen, carbon dioxide, and methane as a non-adsorbed fraction;

(c) passing the non-adsorbed fraction from step (b) through the second stage of the pressure swing adsorption system (M) to separate carbon dioxide and methane as an adsorbed fraction and hydrogen as a non-adsorbed substantially pure product;

(d) stopping the flow of feed mixture into the inlet end of the first stage;

(e) desorbing carbon dioxide and methane from a location intermediate the first and second stages of the pressure swing adsorption system (M) to form a carbon dioxide-rich fraction;

(f) desorbing the carbon dioxide and methane by purging the second stage with hydrogen gas in a countercurrent direction and purging the first stage with carbon monoxide gas in a cocurrent direction and withdrawing the purge effluent gases from a location intermediate the first and second stages of the pressure swing adsorption system (M); and

(g) desorbing carbon monoxide from the first stage of the pressure swing adsorption system (M) to form a substantially pure carbon monoxide product."

IV. The appellant submitted the following arguments in support of his request:

The present only independent claim concerns the embodiment illustrated by original Figures 4 and 5 and is based on the

combination of features of original independent claim 15 with dependent claims 20, 22 and 23.

A PSA method of producing hydrogen and carbon monoxide from a feed mixture comprising moreover carbon dioxide and methane, is known from D1. This method has a drawback in that, because in the first stage of the first PSA plant an adsorbent is used which adsorbs carbon dioxide preferentially over carbon monoxide, a second PSA plant for separating carbon monoxide is needed. The present invention solves this problem and provides a PSA system wherein a second PSA plant is superfluous by, in particular, implementing in the first stage an adsorbent having a greater affinity for carbon monoxide than for hydrogen, carbon dioxide, and methane. Indeed, such type of adsorbents are known from D2. However, starting from the process of D1, with the first plant having the same type of adsorbent in both stages, there is a plurality of ways on which the adsorbent of D2 can be used and, in particular, there is no incentive from D2 to change only one of said both adsorbents, i.e. the one of the bottom stage of the first plant. There is no indication in D2 either about the efficiency of the separation of carbon monoxide in the mentioned adsorbent, so that the skilled person cannot determine whether, by substituting this adsorbent of D2 for the activated carbon adsorbent in a stage of the first PSA plant of D1, the purity of the hydrogen product produced by the second stage of the first PSA plant is not put in jeopardy. Thus, it is only by hindsight that the skilled person, starting from D1, can arrive at the present method by adding the teaching of D2 to that of D1 and, therefore, the subject-matter of the claim involves an inventive step.

Reasons for the Decision

1. The appeal is admissible.

2. *Allowability of the amendments*

It is to be noted that there is no present claim based on original claim 1 and that the appellant has declared that the embodiments illustrated by original Figures 1 to 3 are disclaimed. Present claim 1 results from the combination of the features of original independent claim 15 and original successively dependent claims 20, 22 and 23, which specify the features (d) and (f) and part of the features (b) and (e) of present claim 1. In this respect, it is to be noted that both steps (e) and (f) concern desorbing carbon dioxide and methane from a location intermediate the first and second stages of the pressure swing adsorption system (M); indeed, in agreement with the original description (see Table 3 on page 30 and the table on page 31), present step (e) corresponds to original Step No. 4 "Top/Bottom depressurize" indicated as "Top and bottom beds depressurized from an intermediate location. Depressurized gas recycled to reformer feed gas" and present step (f) corresponds to original Step No. 5 "Top purge Bottom purge" indicated as "Top bed purged with hydrogen. Bottom bed purged with carbon monoxide. Purge effluent removed from intermediate location", respectively. The feature of present claim 1 that the steps are repeated is based on the original description (see in particular page 35, last paragraph, first to sixth line).

Therefore, the European patent application satisfies the requirement of Article 123(2) EPC that it may not be amended in such a way that it contains subject-matter which extends beyond the content of the application as filed.

3. *Clarity*

The Board of appeal is satisfied that the claims define correctly the method of the invention and that, with the amendments to be made in the description and drawings for adaptation to the texts of said claims, they are clear in the sense of Article 84 EPC.

4. *Novelty*

Since the subject-matter of present claim 1 is not known from the state of the art, it is novel in the sense of Article 54 EPC.

5. *Inventive step*

5.1 A method of producing by pressure swing adsorption hydrogen and carbon monoxide products from a feed mixture comprising hydrogen, carbon monoxide, carbon dioxide, and methane, is known from D1 (see page 2, line 35 to page 3, line 5; page 5, lines 40 to 46; page 6, line 28 to page 9, line 18; Figures 1 and 2). The known method, which is acknowledged in the present application (see page 2, last paragraph to page 3, first paragraph), also comprises repeating steps, and in particular the following steps of:

(a) providing a pressure swing adsorption system (122) having a first stage (124; 202) and a second stage (126; 212), wherein the first stage (124; 202) contains an adsorption bed comprising an adsorbent having a greater affinity for, in particular, carbon monoxide than for hydrogen, the second stage (126; 212) contains an adsorption bed comprising (at least) one adsorbent (219, 221) having a greater affinity for carbon dioxide, methane, and carbon monoxide than for hydrogen, and the first and second stages are connected in series and each stage contains an inlet end (120, 203; 205) and an outlet end (128; 205, 207);

(b) passing the feed mixture through the inlet end (120; 203) of the first stage (124; 202) of the pressure swing adsorption system (122) to separate, in particular, hydrogen, as a non-adsorbed fraction;

(c) passing the non-adsorbed fraction from step (b) through the second stage (126; 212) of the pressure swing adsorption system (122) to separate hydrogen as a non-adsorbed substantially pure product.

5.1.1 However, contrary to the presently claimed method, wherein in particular the first stage contains an adsorption bed comprising an adsorbent having a greater affinity for carbon monoxide than for hydrogen, carbon dioxide, and methane, the known method uses as a first stage an adsorbent which has **not** a greater affinity for carbon monoxide than for carbon dioxide.

As derivable from D1 (see in particular page 6, line 53 to page 7, line 15) and as argued by the appellant, in order to obtain carbon monoxide as a pure product, the known method indeed uses a second, separate PSA plant (148) with two stages (150; 152), whereby in particular in the second stage (152) thereof carbon monoxide is preferably adsorbed and carbon monoxide product is obtained by desorbing said stage (152); however, the gas mixture enriched in carbon monoxide that exits the first PSA plant (122) through the intermediate outlet (130) between the first and second stages (124, 126) has to be compressed in the compressor (142) to raise the pressure thereof preferably to about one atmosphere in excess of the pressure of the gas entering the first plant (122). Thus, although the known method indeed comprises some specific steps recited in present claim 1, it does not comprise the specific arrangement of said steps and all the particular features thereof which result from the use of the particular adsorbents with different affinities mentioned here above. Moreover, it is to be noted that the known methods are also distinguished from the presently claimed method under particular aspects concerning further steps resulting from structural differences of the PSA systems in which the known and the presently claimed method are carried out, respectively. For instance, contrary to step (e) of the method of present claim 1, in the method of D1 (see page 6, lines 24 to 27; Figure 1) carbon dioxide and methane are not desorbed from a location intermediate the first (124) and second (126) stages of the pressure swing adsorption system (122), but from the bottom of said PSA system through the conduit (132).

5.2 As derivable from the present application as originally filed (see page 2, last paragraph to page 3, first paragraph; page 6, two last paragraphs) and as convincingly argued by the appellant, this need for two PSA plants, one for producing hydrogen as a product gas and a further one for producing carbon monoxide as an adsorbed product, is a drawback, and an aim of the present invention is to provide an improved, simplified method of producing both hydrogen and carbon monoxide from a feed mixture by a novel combination of pressure swing adsorption methods which in particular minimizes capital cost requirements.

5.3 Yet, at the priority date, an adsorbent having a greater affinity for carbon monoxide than for hydrogen, carbon dioxide, and methane was known from D2, which is indeed cited in the present original application (see page 3, second paragraph). Thus, the person skilled in the art of D1, made aware by D2 (see claim 19; see also column 1, lines 8 to 27 and column 3, lines 30 to 41) of a possibility of obtaining directly carbon monoxide without in particular having to compress the carbon monoxide enriched gas mixture, because there is no necessity for a second PSA plant, could be prompted to use the new adsorbent of D2 in the PSA system of D1, the other method steps having to be arranged and adapted to the new configuration of the PSA apparatus by taking into account, in particular, the indications in D1 (see page 7, lines 9 to 13; page 8, lines 52 to 55; Figures 1 and 2) about desorbing and purging adsorbed gas and the generally known advantages of each of the steps of an adsorption process.

5.4 In this respect, the appellant has convincingly argued as follows:

In the process of D1 (see page 6, lines 34 to 39; page 8, lines 6 to 13; Figures 1 to 3), the adsorbent in both beds (124; 126) of the first PSA plant (122) preferentially adsorbs in the order: carbon dioxide, carbon monoxide and hydrogen; in accordance with this indication, the part (210) in the bottom stage (202) and the bottom part (219) of the second, upper stage (212) are both made of activated carbon adsorbent; the upper part (221) of said upper stage (212), which is made of zeolite molecular sieve adsorbent, also adsorbs in particular carbon dioxide. Moreover, in the process of D1 (see page 6, line 53 to page 7, line 15; see also Figure 5 and the corresponding text), there is a further PSA plant (148) comprising a first stage (150) wherein constituents in the gas mixture enriched in carbon monoxide that exits the first PSA plant (122) are submitted to a selective adsorption to produce a gas mixture essentially of carbon monoxide and hydrogen, and a second stage (152) wherein carbon monoxide is adsorbed to produce a gas mixture rich in hydrogen, which is sent again to the first PSA plant (122).

Thus, starting from the process known from D1, with the first plant having the same type of adsorbent in both stages, there is a plurality of ways of implementing the adsorbent of D2 and, in particular, there is no incentive from D2 to change only one of said both adsorbents, i.e. the one of the bottom stage (202). There is no indication in D2 either about the efficiency of the separation of carbon monoxide in the mentioned adsorbent, so that the

skilled person could not determine whether, by substituting this adsorbent of D2 for the activated carbon adsorbent in the bottom stage, or in both the bottom and the upper stage of the first PSA plant of D1, even by keeping the zeolite at the top of the upper stage, the purity of the hydrogen product produced by said second stage of the first PSA plant would be put in jeopardy. Thus, it is only by hindsight that the skilled person, starting from D1, could arrive at the presently claimed method by implementing the teaching of D2.

5.5 The following is to be noted with respect to the argumentation of the appellant:

Starting from D1, the skilled person could indeed get aware that
(1) the two step PSA method of D1 is complicated, that
(2) an adsorbent having different adsorbing affinities, for instance adsorbing preferentially carbon monoxide over carbon dioxide and not the contrary, can lead to a different PSA system, and that
(3) moreover the adsorbent of D2 is adsorbing preferentially carbon monoxide over carbon dioxide.

However, there is a plurality of different beds wherein the skilled person could substitute the adsorbent of D2 for some of the adsorbents of the system of D1, or even add the adsorbent of D2 to those of D1. Moreover, for each of said specific implementation of the adsorbent of D2, there are modifications of the system of D1 necessary for adapting said system while keeping at least the capability mentioned in D1 (see page 2, lines 35 to 37; page 7, lines 19 to 20)

of obtaining both hydrogen and carbon monoxide products of substantial purity. In this respect, it is to be noted that there is not much information derivable from D2 about the capabilities of the adsorbent mentioned therein. More importantly, it is to be noted that the presently claimed method uses an adsorbent of the type mentioned in D2 which has inverted properties with respect to those of D1, i.e. which adsorbs preferentially carbon monoxide over carbon dioxide and not the contrary, and that this adsorbent is to be used in steps (a) and (b), i.e. in the initial steps of the method, wherein the feed mixture is passed through the inlet end of the first stage of the PSA system (M) to separate carbon monoxide as an adsorbed fraction and hydrogen, carbon dioxide, and methane as a non-adsorbed fraction. This inversion of the adsorbing capability already in the initial step of the method as compared with D1, with all the adaptations resulting thereof, leads to the necessity of re-designing the whole system from the beginning and is indeed an indication that, to arrive at the presently claimed method by starting from D1 and combining with D2, hindsight is necessary.

Therefore, the subject-matter of present claim 1 involves an inventive step in the sense of Article 56 EPC and, thus, a patent can be granted (Articles 52(1) and 97(2) EPC).

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of claims 1 to 5 presented at the oral proceedings of 31 January 1996, with the description and drawings to be adapted.

The Registrar:The Chairman:

P. Martorana E. Turrini