

Title (en)

METHOD FOR PRODUCING FORMIC ACID BY REACTING CARBON DIOXIDE WITH HYDROGEN

Title (de)

VERFAHREN ZUR HERSTELLUNG VON AMEISENSÄURE DURCH UMSETZUNG VON KOHLENDIOXID MIT WASSERSTOFF

Title (fr)

PROCÉDÉ DE PRODUCTION D'ACIDE FORMIQUE PAR RÉACTION DE DIOXYDE DE CARBONE AVEC DE L'HYDROGÈNE

Publication

EP 2763950 A2 20140813 (DE)

Application

EP 12766684 A 20121002

Priority

- EP 11184297 A 20111007
- EP 2012069458 W 20121002
- EP 12766684 A 20121002

Abstract (en)

[origin: WO2013050367A2] The invention relates to a method for producing formic acid, comprising the following steps: (a) reacting, in a homogeneously catalyzed manner, a reaction mixture (Rg) containing carbon dioxide, hydrogen, at least one polar solvent selected from the group comprising methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 2-methyl-1-propanol, and water, and at least one tertiary amine of general formula (A1) $\text{NR}_1\text{R}_2\text{R}_3$, in which R1, R2, and R3 represent independently of one another an unbranched or branched, acyclic or cyclic, aliphatic, araliphatic, or aromatic group having 1 to 16 carbon atoms, wherein individual carbon atoms can also be substituted independently of one another with a hetero group selected from the groups -O- and >N- and two or all three residues can also be bonded to one another to form a chain comprising at least four atoms, in the presence of at least one coordination catalyst, which contains at least one element selected from groups 8, 9, and 10 of the periodic system, in a hydrogenation reactor in order to obtain, optionally after adding water, a two-phase hydrogenation mixture (H) containing an upper phase (O1), which contains the at least one coordination catalyst and the at least one tertiary amine (A1), and a lower phase (U1), which contains the at least one polar solvent, residues of the at least one coordination catalyst, and at least one formic acid/amine adduct of general formula (A2), $\text{NR}_1\text{R}_2\text{R}_3 \cdot x \text{HCOOH}$, in which x is in the range of 0.4 to 5 and R1, R2, R3 have the meanings stated above; (b) processing the hydrogenation mixture (H) obtained in step (a) according to one of the following steps: (b1) phase-separating the hydrogenation mixture (H) obtained in step (a) in a first phase-separating device into the upper phase (O1) and the lower phase (U1), or (b2) extracting the at least one coordination catalyst from the hydrogenation mixture (H) obtained in step (a) in an extraction unit with an extracting agent containing the at least one tertiary amine (A1) in order to obtain a raffinate (R1) containing the at least one formic acid/amine adduct (A2) and the at least one polar solvent and an extract (E1) containing the at least one tertiary amine (A1) and the at least one coordination catalyst, or (b3) phase-separating the hydrogenation mixture (H) obtained in step (a) in a first phase-separating device into the upper phase (O1) and the lower phase (U1) and extracting the residues of the at least one coordination catalyst from the lower phase (U1) in an extraction unit by means of an extracting agent containing the at least one tertiary amine (A1) in order to obtain a raffinate (R2) containing the at least one formic acid/amine adduct (A2) and the at least one polar solvent and an extract (E2) containing the at least one tertiary amine (A1) and the residues of the at least one coordination catalyst; (c) separating the at least one polar solvent from the lower phase (U1), from the raffinate (R1), or from the raffinate (R2) in a first distillation device in order to obtain a distillate (D1) containing the at least one polar solvent, which is fed back into the hydrogenation reactor in step (a), and a two-phase bottom mixture (S1) containing an upper phase (O2), which contains the at least one tertiary amine (A1), and a lower phase (U2), which contains the at least one formic acid/amine adduct (A2); (d) optionally processing the bottom mixture (S1) obtained in step (c) by phase separation in a second phase-separating device into the upper phase (O2) and the lower phase (U2); (e) cleaving the at least one formic acid/amine adduct (A2) contained in the bottom mixture (S1) or optionally in the lower phase (U2) in a thermal cleaving unit in order to obtain the at least one tertiary amine (A1), which is fed back to the hydrogenation reactor in step (a), and formic acid, which is discharged from the thermal cleaving unit, wherein carbon monoxide is added to the lower phase (U1), the raffinate (R1), or the raffinate (R2) directly before and/or during step (c) and/or carbon monoxide is added to the bottom mixture (S1) or optionally the bottom phase (U2) directly before and/or during step (e).

IPC 8 full level

C07C 51/02 (2006.01); **B01J 31/24** (2006.01); **C07C 51/41** (2006.01); **C07C 51/48** (2006.01); **C07C 53/02** (2006.01); **C07C 53/06** (2006.01)

CPC (source: CN EP)

B01J 31/0202 (2013.01 - EP); **B01J 31/0237** (2013.01 - EP); **B01J 31/24** (2013.01 - EP); **B01J 31/2414** (2013.01 - EP); **C07C 51/00** (2013.01 - CN); **C07C 51/02** (2013.01 - CN EP); **C07C 51/41** (2013.01 - EP); **C07C 51/42** (2013.01 - CN); **C07C 51/48** (2013.01 - EP); **B01J 2231/625** (2013.01 - EP); **B01J 2531/821** (2013.01 - EP); **B01J 2531/98** (2013.01 - EP)

Citation (search report)

See references of WO 2013050367A2

Cited by

WO2016024293A1

Designated contracting state (EPC)

AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO RS SE SI SK SM TR

DOCDB simple family (publication)

WO 2013050367 A2 20130411; **WO 2013050367 A3 20131003**; BR 112014008338 A2 20170418; CA 2851175 A1 20130411; CN 103974927 A 20140806; EP 2763950 A2 20140813; IN 3122DEN2014 A 20150522; JP 2014530216 A 20141117; KR 20140074373 A 20140617; RU 2014118032 A 20151120; SG 11201400997T A 20140926

DOCDB simple family (application)

EP 2012069458 W 20121002; BR 112014008338 A 20121002; CA 2851175 A 20121002; CN 201280059095 A 20121002; EP 12766684 A 20121002; IN 3122DEN2014 A 20140421; JP 2014533856 A 20121002; KR 20147011873 A 20121002; RU 2014118032 A 20121002; SG 11201400997T A 20121002