

Title (en)

METHOD FOR PRODUCING FORMIC ACID BY REACTING CARBON DIOXIDE WITH HYDROGEN

Title (de)

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Title (fr)

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

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Application

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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 R_1 , R_2 , and R_3 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 R_1 , R_2 , R_3 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).

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Cited by

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