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**Datasheet for the decision  
of 30 May 2011**

**Case Number:** T 0967/08 - 3.3.01

**Application Number:** 01992145.1

**Publication Number:** 1355911

**IPC:** C07F 7/02

**Language of the proceedings:** EN

**Title of invention:**

Nanosized copper catalyst precursors for the Direct Synthesis  
of trialkoxysilanes

**Applicant:**

GENERAL ELECTRIC COMPANY

**Opponent:**

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**Headword:**

Nanosized copper/GENERAL ELECTRIC

**Relevant legal provisions:**

EPC Art. 84, 123(2)

**Relevant legal provisions (EPC 1973):**

-

**Keyword:**

"Main request and auxiliary request II: clarity of the claims  
(no) - the parameter "average particle size" renders the claim  
unclear as neither the type of average nor a method for  
determining it is indicated in the claims"

"Auxiliary request I - contravening the requirements of  
Article 123(2) EPC"

**Decisions cited:**

T 1819/07, T 0728/98

**Catchword:**

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Case Number: T 0967/08 - 3.3.01

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.01  
of 30 May 2011

**Appellant:** GENERAL ELECTRIC COMPANY  
1 River Road  
Schenectady  
NY 12345 (US)

**Representative:** Wibbelmann, Jobst  
Wuesthoff & Wuesthoff  
Patent- und Rechtsanwälte  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 29 October 2007  
refusing European patent application  
No. 01992145.1 pursuant to Article 97(1) EPC  
1973.

**Composition of the Board:**

**Chairman:** P. Ranguis  
**Members:** C. M. Radke  
L. Bühler

## Summary of Facts and Submissions

- I. The applicant filed an appeal against the decision of the examining division to refuse European patent application No. 01 992 145.1.
- II. The following document was cited *inter alia* during the examination proceedings:
- (D1) US-A-5 362 897.
- III. The examining division deemed that
- the introduction of the features of original claim 6 into claim 1 contravened the requirements of Article 123(2) EPC;
  - claim 8 was unclear, as the addition of an alcohol to the copper-activated silicon was mandatory according to claim 1, whereas dependent claim 8 also permitted the addition of the silicon to the alcohol;
  - the subject-matter of claims 20-21 contravened the requirements of Article 83 EPC, as the conditions for measuring the surface area were not indicated; and
  - the subject-matter of the claims was not inventive in view of document (D1).
- IV. The refusal was based on claims 1-21 filed by letter dated 24 August 2007, the only independent claim reading as follows:
- "1. A process for the Direct Synthesis of trialkoxysilane of formula  $\text{HSi}(\text{OR})_3$  wherein R is an alkyl group containing from 1 to 6 carbon atoms

using a member selected from the group consisting of nanosized copper, nanosized copper oxides, nanosized copper chlorides, other nanosized copper salts, and mixtures thereof as sources of catalytic copper, having an average particle size in a range from about 0.1 to about 600 nanometers, said process comprising:

- (a) forming a reaction mixture comprising a thermally stable solvent, silicon metal, a catalytically effective amount of said nanosized copper catalyst precursor;
- (b) agitating and heating this mixture to form copper-activated silicon *in situ* and injecting into said reaction mixture an alcohol to react with said copper-activated silicon to produce said trialkoxysilane; and
- (c) recovering said trialkoxysilane from the reaction product."

V. This decision on the appeal is based on the following sets of claims filed by letter dated 2 May 2011:

- Claims 1-18 of the main request;
- claims 1-18 of auxiliary request I, and
- claims 1-5 of auxiliary request II (which are identical to claims 1-5 of the main request).

- (a) Claim 1 of the main request and claim 1 of auxiliary request II read as follows:

"1. A process for the Direct Synthesis of trialkoxysilane of formula  $\text{HSi}(\text{OR})_3$  wherein

R is an alkyl group containing from 1 to 6 carbon atoms using a member selected from the group consisting of nanosized copper, nanosized copper oxides, nanosized copper chlorides, other nanosized copper salts, and mixtures thereof, having an average particle size in a range from 0.1 to 100 nanometers, as sources of catalytic copper, said process comprising:

- (a) forming a reaction mixture comprising a thermally stable solvent, silicon metal, a catalytically effective amount of said nanosized copper catalyst precursor;
- (b) agitating and heating this mixture to form copper-activated silicon *in situ* and injecting into said reaction mixture an alcohol to react with said copper-activated silicon to produce said trialkoxysilane; and
- (c) recovering said trialkoxysilane from the reaction product."

- (b) Claim 1 of auxiliary request I differs from claim 1 of the main request in that the expression "having an average particle size in a range from 0.1 to 100 nanometers" was replaced by "having a range of particle sizes between 0.1 to 100 nanometers inclusive".

VI. The Board issued a communication on 18 February 2011 in which it *inter alia* raised objections as to the clarity of the claims then on file caused by the parameter "average particle size" (see point 3.1 of the communication). Due to this lack of clarity the Board doubted whether the subject-matter of the claims then

on file was novel in view of the disclosure of document (D1) (see point 4 of the communication). The Board enclosed the following document with this communication:

(D4) N. Stanley-Wood, Particle Size Analysis: Introduction, pages 1-14, Encyclopedia of Analytical Chemistry, John Wiley & Sons Ltd © 2000, published online on 15 September 2006.

On 25 May 2011, the Board provided the appellant with the following document by e-mail:

(D7) DIN ISO 9276-2 dated February 2006, Beuth Verlag GmbH, Berlin/DE, 17 pages.

VII. The appellant argued that the claims of the main request and of auxiliary request II were clear; document (D4) confirmed that the person skilled in the art would determine the average particle size according to the standard (D7) which gave a unique definition for said parameter. The different methods for determining average particle size lead to similar results.

The claims of auxiliary request I met the requirements of Article 123(2) EPC as the amendments were based on pages 41 and 42 of the application as filed, in particular Table V on page 41.

VIII. The appellant requested that the decision under appeal be set aside and the case be remitted to the first instance with the order to grant a patent based on the claims of the main request, auxiliary request I or auxiliary request II and amended pages 1, 44, 45 and 47 submitted with the letter dated 2 May 2011.

IX. At the end of the oral proceedings the chairman announced the decision of the Board.

### **Reasons for the Decision**

1. The appeal is admissible.

2. *Clarity of the claims of the main request and of auxiliary request II*

2.1 The Board mentioned in its communication that document (D4) stated the following (see the second paragraph of chapter 3.2 on page 13):

"The general description of an average particle size within a powder is thus dependent upon both the type of size distributions and the method of calculation from moments of the particle size mean."

The appellant did not provide any evidence for its statement that the values of average particle sizes determined by different methods were similar.

Moreover, it is known that these values may differ considerably. Reference is made to T 1819/07 of 15 March 2011, where the following is stated under point 3.2 of the reasons:

"... average particle sizes (more precisely: mean particle diameters) such as the arithmetic mean diameter (or **count** mean diameter)  $\bar{d}$ , the **volume or mass** mean diameter  $\bar{d}_v$  and the mean surface **area** diameter  $\bar{d}_s$



are among the most commonly used quantities for describing the average diameter of a particle population (...). The values of the average particle sizes  $\bar{d}$ ,  $\bar{d}_v$ , and  $\bar{d}_s$  differ for any particle size distribution (...).

...

Hence, there are different methods for determining the average particle size yielding values for the same particle distribution and these generally differ, under particular conditions by one or two orders of magnitude."

2.2 The appellant referred to the first complete sentence in the right-hand column on page 13 of document (D4) which reads as follows:

"Equations for the calculations of average particle sizes or average particle diameters from a given particle size distribution are defined in ISO 9276-2<sup>(39)</sup> to give a unique definition of average size, derived from the moments of a size distribution."

The respective standard (D7) does not, however, give one definition of average particle size but rather provides equations for calculating the different kinds of averages (see document (D7), third page (page 1 of ISO 9276-2:2001), namely the arithmetic averages (chapter 5.1), weighted averages (chapter 5.2) and further average particle diameters (Annex B); see page 5, fourth to second line from the bottom, which refer to length, surface and volume averages).

- 2.3 For these reasons the Board concludes that
- the person skilled in the art would equally consider several methods when determining average particles sizes, and
  - these different methods yield considerably different values.

Claim 1 refers to a range of average particle sizes without indicating which type of average is to be taken or by which method it is to be determined.

Hence, for a given particle size distribution, the average particle size determined by one method may be within the range indicated in claim 1 whereas it may be outside said range when determined by another method. Therefore, this feature is unclear.

- 2.4 "A claim comprising an unclear technical feature, ..., entails doubts as to the subject-matter covered by that claim. This applies all the more if the unclear feature is essential with respect to the invention in the sense that it is designed for delimiting the subject-matter claimed from the prior art, thereby giving rise to uncertainty as to whether or not the subject-matter claimed is anticipated." (T 728/98, OJ EPO 2001, 319, point 3.1 of the reasons).

The latter applies to the feature "average particle size" in the present case (see the novelty objection mentioned under point VI above).

- 2.5 For these reasons claim 1 of the main request and of auxiliary request II is unclear.

2.6 The Board may only decide on a request as a whole. Therefore, the main request and auxiliary request II are refused.

3. *Amendments in the claims of auxiliary request I*

3.1 Claim 1 of the main request referred to "an **average** particle size in a range from 0.1 to 100 nanometers" as disclosed in claim 8 as originally filed. In claim 1 of auxiliary request I, this expression was replaced by "having a **range of particle sizes** between 0.1 to 100 nanometers inclusive" (emphasis added by the Board). Hence, according to this claim, not only the average particle size but also the size of every single particle must be within the given range.

3.2 The appellant referred to pages 41 and 42 of the description as originally filed as a basis for this amendment. The only information on these pages relating to particle sizes is Table V on page 41:

**Table V**  
**Nanosized Copper And Copper Oxides Used In  
Examples 4A to 4D**

<b>EXAMPLE</b>	<b>HYDROCARBON</b>	<b>NANOPHASES</b>	<b>PARTICLE SIZE</b>
4A	CARNATION® 70	Cu <sub>2</sub> O major, CuO minor	1 to 15 nm
4B	CARNATION® 70	Cu <sub>2</sub> O major, CuO minor	1 to 15 nm
4C	ISORCHEM® 113	Cu <sub>2</sub> O major, CuO minor	20 to 100 nm
4D	SIRENE® X12L	Cu <sub>2</sub> O major, Cu minor	20 to 50 nm

The lowest particle size mentioned in this table is one nanometer (1 nm). Therefore, it is evident that this table does not disclose nanosized copper and copper oxides having a range of particle sizes between **0.1** to 100 nanometers inclusive. Nor does any other part of

the application as filed relating to the invention disclose said range.

3.3 Hence, present claim 1 as amended contains subject-matter which extends beyond the content of the application as filed, contrary to the requirements of Article 123(2) EPC.

3.4 In view of this conclusion the Board does not need to assess whether or not each range of particle sizes given in said table is so closely linked to the composition of the respective nanophase that its generalisation to any composition of nanophases defined in claim 1 contravenes the requirements of Article 123(2) EPC.

3.5 For the reasons mentioned under points 3.2 and 3.3 above, auxiliary request I is refused.

## **Order**

### **For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:

M. Schalow

P. Ranguis