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**Datasheet for the decision  
of 15 March 2011**

**Case Number:** T 1819/07 - 3.3.01

**Application Number:** 01985985.9

**Publication Number:** 1337537

**IPC:** C07F 7/16

**Language of the proceedings:** EN

**Title of invention:**

Rochow-Muller direct synthesis using copper catalyst precursors with particle dimensions in the nano-region

**Patentee:**

GENERAL ELECTRIC COMPANY

**Opponent:**

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**Headword:**

Nanosized catalyst/GENERAL ELECTRIC

**Relevant legal provisions:**

EPC Art. 84

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

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**Keyword:**

"Clarity of the claims (no) - the parameter "average particle size" renders the claims unclear as neither the type of average (volume, surface, number) nor a method for determining it is indicated in the claims"

**Decisions cited:**

T 0908/04

**Catchword:**

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Case Number: T 1819/07 - 3.3.01

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.01  
of 15 March 2011

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

**Representative:** Wibbelmann, Jobst  
Wuesthoff & Wuesthoff  
Patent- und Rechtsanwälte  
Schweigerstraße 2  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 31 May 2007  
refusing European patent application  
No. 01985985.9 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. 01 985 985.9.
- II. The following documents were cited during the examination proceedings:
- (D1) EP-A-0 784 056
  - (D2) WO-A-85/05 047
  - (D3) US-A-5 759 230
  - (D4) US-A-4 450 282.
- III. The examining division decided that the claims lacked unity of invention *a posteriori* in view of document (D1).
- IV. The refusal was based on claims 1 to 18 filed with the letter dated 30 March 2007, the only independent claim reading as follows:
- "1. A method for Direct Synthesis of organohalosilanes comprising the steps of :
- providing a silicon metal;
  - providing an organohalide of formula RX wherein R is a saturated or unsaturated aliphatic or aromatic hydrocarbon radical having from 1 to 10 carbon atoms, and X is a halogen;
  - providing a copper catalyst precursor selected from the group consisting of copper metal, copper (I) oxide, copper (II) oxide, copper (I) chloride, copper (II) chloride, copper (I) carboxylates, copper (II) carboxylates, other copper salts, and

mixtures thereof, said copper catalyst precursor **having an average particle size from about 0.1 to about 600 nanometers;**

providing one or more promoters; and  
reacting said silicon metal, said organohalide and said copper catalyst precursor for a time and temperature to effectuate D/T selectivity (gravimetric ratio of  $R_2SiX_2/RSiX_3$ ) of greater than 10." (Emphasis added by the Board).

V. This decision is based on the following sets of claims filed with the letter dated 11 February 2011:

Claims 1 to 18 of the Main Request,  
claims 1 to 17 of the First Auxiliary Request, and  
claims 1 to 4 of the Second Auxiliary Request.

Each of these sets of claims contains claim 1 as the only independent claim.

(a) Claim 1 of the Main Request only differs from the claim 1 cited under point IV above in that the feature "having an average particle size from about 0.1 to about 600 nanometers" was replaced by **"having an average particle size from 0.1 to 600 nanometers"**.

(b) Claim 1 of the First Auxiliary Request reads as follows:

"1. A method for Direct Synthesis of organohalosilanes comprising the steps of :  
providing a slurry of silicon,

an organohalide of formula RX wherein R is a saturated or unsaturated aliphatic or aromatic hydrocarbon radical having from 1 to 10 carbon atoms, and X is a halogen, a copper catalyst precursor selected from the group consisting of copper metal, copper (I) oxide, copper (II) oxide, copper (I) chloride, copper (II) chloride, copper (I) carboxylates, copper (II) carboxylates, other copper salts, and mixtures thereof, said copper catalyst precursor **having an average particle size from 0.1 to 600 nanometers**, and one or more promoters in a thermally stable liquid solvent; and reacting said silicon, said organohalide and said copper catalyst precursor in said slurry for a time and temperature to effectuate D/T selectivity (gravimetric ratio of  $R_2SiX_2/RSiX_3$ ) of greater than 10." (Emphasis added by the Board).

- (c) Claim 1 of the Second Auxiliary Request reads as follows:

"1. A method of controlling a Direct Synthesis for making an organohalosilane comprising the steps of:  
providing a silicon metal;  
providing one or more copper catalyst precursors **having an average particle size of less than 600 nanometers**;  
providing one or more promoters selected from the group consisting of zinc, cadmium, antimony, phosphorus, arsenic, lanthanides, alkali metal halides, tin, related compounds, and mixtures

thereof, having an average particle size of less than 500 nanometers, wherein a Zn/Sn gravimetric ratio is 12 to 60, and a Zn/Cu gravimetric ratio is 0.04 to 0.2;

heating said silicon metal, said one or more copper catalyst precursors, and said one or more promoters;

forming copper-silicon intermetallics for reaction with an organohalide; and

maintaining the zinc to tin ratio during the Direct Synthesis wherein D/T selectivity (gravimetric ratio of  $R_2SiX_2/RSiX_3$ ) for a dialkyldihalosilane is greater than 10." (Emphasis added by the Board).

VI. The Board issued a communication in which it *inter alia* raised objections as to the clarity of the claims as far as the parameter "average particle size" is concerned. The Board enclosed the following document with this communication:

(D5) Kirk-Othmer, Encyclopedia of Chemical Technology, 4th edn., vol. 22, John Wiley & Sons, Inc., New York/US 1997, pages 256-278.

VII. The Appellant argued that the claims were clear as the method for determining the average particle size was incorporated into the specification by reference to an application published as

(D6) US-B-7 087 100.

It considered the method described in this document to be the most common one by which the person skilled in the art would determine the average particle size.

- VIII. During the oral proceedings, the Board stated
- that the claims must be clear as such;
  - that the present application referred to the U.S. patent application on which the patent (D6) is based only as far as the preparation of the nanosized copper and copper oxides is concerned; and
  - that documents (D1) to (D4) and (D6) taught different methods of determining the average particle size, whereas document (D5) taught that these different methods yield different values.

- IX. The Appellant requested that the decision under appeal be set aside and that the case be remitted to the department of first instance with the order to continue examination based on the Main Request, the First Auxiliary Request or the Second Auxiliary Request.

These requests are based on the respective sets of claims and on page 38 of the description, all filed with the letter dated 11 February, 2011 (see under point V above).

- X. 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. Article 123(2) EPC

In view of the outcome of this decision it is not necessary to decide whether or not the present claims satisfy the requirements of Article 123(2) EPC.

3. Clarity of the claims

- 3.1 Claim 1 of the Main Request and of both Auxiliary Requests characterises the copper catalyst precursor by indicating a range for the "average particle size" of from 0.1 to 600 nanometers or of less than 600 nanometers.

- 3.2 Document (D5) states that 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 (see page 257, the first sentence under the heading "Distribution Averages", and formulae (1), (2) and (3) on pages 257-258). The values of the average particle sizes  $\bar{d}$ ,  $\bar{d}_v$ , and  $\bar{d}_s$  differ for any particle size distribution (see equation (4) on page 258).

"It is not unusual for a polydispersed particle population to exhibit a diameter of average mass as being one or two orders of magnitude larger than the



arithmetic mean of the diameters." (see page 258, the penultimate sentence of the first paragraph under equation (3)).

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

- 3.3 The Appellant considered the method described in document (D6) to be the most common one by which the person skilled in the art would determine the average particle size (see point VII above).
- 3.3.1 This document mentions that the "Determination of particle size may be done by transmission electron microscopy (TEM) or high resolution scanning electron microscopy (HRSEM)." (see column 6, lines 23-25).
- 3.3.2 The present application deals with the Direct Synthesis of organohalosilanes, i.e. the copper catalysed reaction of silicon with organohalides (see page 1, lines 10-16 of the application as published). Of the prior art documents cited by the Examining Division, documents (D1), (D2) and (D4) also deal with said Direct Synthesis (see (D1), page 2, lines 3-12; (D2), page 1, lines 3-8); (D4), column 1, lines 6-22).

Document (D1) states that the "mean particle size ... indicates a median diameter known as  $d_{P50}$  corresponding to 50% on a **volume** basis cumulative distribution curve ..." (see page 4, lines 26-27; emphasis added by the Board).

According to document (D2) "... an average particle size means the **mass** median particle size as measured with ..." (see page 7, lines 11-15; emphasis added by the Board).

Document (D4) considers it "desirable that the **area** mean diameter of the particles" be within a certain range (see column 9, lines 1-13; emphasis added by the Board).

Hence, there is no reason to assume that there is a most common method of determining the average particle size in the field of the Direct Synthesis of organohalosilanes. Moreover, the values determined by calculating the **volume or mass average** will differ from those taking the **area** average (see under point 3.2 above).

3.4 Finally, the Appellant argued that the claims were clear as the method for determining the average particle size was disclosed in U. S. patent (D6) as the respective patent application was referred to in the application as filed (see under point VII above).

3.4.1 On the one hand, Article 84 requires that the claims shall be clear. "This implies that the claims must be clear in themselves when being read with the normal skills, but not including any knowledge derived from the description of the patent application ..."  
(T 0908/04 of 15 February 2006, point 3.5 of the reasons). Therefore, a reference in the description can generally not render an otherwise unclear claim clear.

3.4.2 On the other hand, the Appellant relies on references to the US patent application on which the patent (D6) was granted. Said references are found in the application as published on page 14, lines 1-8, and on page 30, lines 12-16.

These references are the following (emphasis added):

"It is preferred that **the nanosized copper and copper oxides** used in the present invention **are prepared by** the methods taught ..., and, most preferably, by the method taught in co-pending patent application entitled PREPARATION OF NANOSIZED COPPER AND COPPER COMPOUNDS, Attorney Docket No. 0066-OS, filed on even date herewith."

" **Nanosized copper (I) oxide was prepared by** thermal decomposition of KOCIDE<sup>®</sup> Cu(OH)<sub>2</sub> in NALKYLENE<sup>®</sup> 500 as taught in co-pending patent application entitled PREPARATION OF NANOSIZED COPPER AND COPPER COMPOUNDS, Attorney Docket No. 0066-OS."

This means that the nanosized particles could be made according to the method disclosed in this document, **not** that the **average particle size** is to be determined according the method described in said U. S. patent application.

For this reason, the methods for determining the average particle size as disclosed in document (D6) in column 6, lines 23-25 (or, more precisely, on page 9, lines 7-8 of the U. S. patent application bearing the Attorney Docket No. 0066-OS) do not form part of the disclosure of the present application as filed. Hence,

these methods can neither establish clarity of the claims nor form the basis for an amendment which might render the claims clear.

3.5 Therefore, the parameter "average particle size" in claim 1 of the Main Request and of the First and Second Auxiliary Requests renders these claims unclear. As claim 1 is the only independent claim in each of these requests, the parameter "average particle size" forms part of a mandatory feature in each claim of these requests, thus rendering all these claims unclear.

3.6 Hence, the claims of the Main Request and of the First and Second Auxiliary Requests do not meet the requirement of Article 84 EPC that the claims shall be clear. As no other set of claims was filed as a basis for a further auxiliary request, the Board had to dismiss the appeal.

## **Order**

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

The appeal is dismissed.

The Registrar:

The Chairman:

M. Schalow

P. Ranguis