

EUROPEAN QUALIFYING EXAMINATION 2010

Paper B(Ch)

Chemistry

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Annex 1 (Patent Application)

Leads for Coloured Pencils

5 [001] The application relates to leads for coloured pencils and in particular to leads suitable for making pencils which leave erasable marks on paper.

[002] Traditional black pencils consist of a lead adhesively bonded in a wooden sheath. Leads for black pencils are made from a blend of kaolin and graphite, which is extruded
10 and fired at a temperature of 950-1200°C. The lead is then impregnated with a wax. Finally the lead is bonded with the sheath to make the pencil. The marks made by such pencils can readily be erased from paper.

[003] Coloured pencils are made by a different method. A metal oxide, a binder, a
15 pigment, lubricants and dispersants are mixed and extruded to form the lead. Coloured leads are generally only heated to temperatures below 150°C, since the pigments used are unable to withstand higher temperatures. Typically the marks made on paper with coloured pencils can only be erased by prolonged rubbing of the paper with an eraser. Sometimes the paper will become damaged before the marks can be removed. There is
20 thus a great need in the art for coloured pencils which will produce easily erasable marks.

[004] The present inventors have carefully studied the differences between coloured and black pencils. They came to the conclusion that the mark made by a black pencil is
25 confined to the surface of the paper, whereas the mark made by coloured pencils penetrates into the structure of the paper. The inventors believe that this is due to the fact that coloured leads are softer. The inventors have thus proposed a harder lead. This is achieved by using hexagonal boron nitride or mica in the lead. It is also believed that the layered structure of hexagonal boron nitride or mica increases the likelihood that the
30 pencil mark will be retained at the surface of the paper.

[005] The pencil lead compositions of the invention comprise a binder, a dispersant, a pigment and a lubricant and are characterised by the fact that they contain at least 20% by weight of hexagonal boron nitride or mica. A preferred composition for a pencil lead in accordance with the invention consists of (all the percentages in the application are by weight):

5
hexagonal boron nitride or mica: 25-45%
metal oxide up to 60%
binder: 5-10%
10 dispersant: 0.5-2.0%
lubricant: 10-25%
pigment: 0.2-30%
up to 5% optional components,
with the proviso that the sum of the metal oxide, hexagonal boron nitride and mica
15 contents is 40-85%.

[006] The most preferred composition is:

20 hexagonal boron nitride or mica: 25-35%
metal oxide: 15-45%
binder: 5-10%
dispersant: 1.0-1.5%
lubricant: 10-20%
pigment: 0.2-30%
25 with the proviso that the sum of the metal oxide, hexagonal boron nitride and mica
contents is 50-70%.

[007] Boron nitride and mica are comparatively expensive and it is thus preferred that a metal oxide such as aluminium oxide or titanium dioxide is also present. Pencil leads
30 containing 25-45% hexagonal boron nitride or mica make marks which are particularly
easy to erase.

[008] The binder may be polyvinyl alcohol, a modified starch, a cellulose derivative or a mixture of these binders. The binder is preferably a cellulose ether and may be selected from methyl cellulose or hydroxypropyl cellulose or hydroxypropyl ethyl cellulose. If the binder is a cellulose ether, 0.1-0.5% of a cross-linking agent may be added to the composition. The cross-linking agent is typically an aldehyde or a diisocyanate. Suitable aldehydes include benzaldehyde, glutaraldehyde and glyoxal. A suitable diisocyanate is methyl phenyl diisocyanate. The cross-linking agent reacts with the binder and increases the hardness of the lead.

[009] The dispersant may be selected from the low toxicity dispersants commercially available and is typically a sorbitan ester.

[010] The lubricant preferably contains a fatty acid with at least 16 carbon atoms or a metal salt of the fatty acid. A particularly preferred fatty acid lubricant is stearic acid. The preferred metal salts are calcium stearate and/or zinc stearate. Up to half of the lubricant may be a wax. The use of a lubricant containing a wax significantly reduces wear on the machinery used to make the leads, but the use of wax in combination with a hexagonal boron nitride or mica filler results in a higher percentage of leads breaking. Leads that break during the manufacturing process have to be discarded and represent a significant additional cost.

[011] The pigments used are standard and are selected in accordance with the colour that the pencil should have. Optional components include bittering agents, fragrances and preservatives. These components are preferably not used.

[012] The process used to make the pencil leads involves kneading together all of the ingredients with a minimum of water. The paste produced is then extruded to make the leads. The extruded leads are dried at a temperature of 50-150°C for 1-4 hours and once they have cooled, are then ready to be incorporated into pencils using a conventional process. If the binder is to be cross-linked then the temperature during the drying step must be at least 110°C in order to ensure that the cross-linking reaction proceeds to completion.

Examples

10

[013] Example 1

A pencil lead composition was made consisting of:

sum of aluminium oxide + hexagonal boron nitride (sum = 65%)

15 binder: methyl cellulose 7%

dispersant: sorbitan ester 1%

lubricant: stearic acid 17%

pigment: Green No. 7 10%

20 [014] The lead was made by: kneading together the ingredients with a minimum amount of water to form a paste, extruding leads from the mixture and drying the leads at 120°C for 2 hours. The leads produced were made into pencils using a conventional process. The amount of hexagonal boron nitride was varied. The writing properties were tested in accordance with the standard 4B test. The erasing of marks was tested with a commercial eraser and rated 1-5 with a value of 1 indicating that the mark is difficult to
25 erase and a value of 5 that a mark is very easy to erase. The percentage of leads which had to be rejected due to breakage or warping was also evaluated.

Percentage of hexagonal boron nitride in lead	Writing properties	Eraser Test	Percentage of Rejects
0	good	1	0.5
15	good	1	1
25	good	4	1
35	good	4	1
45	good	4	1.5
55	good	3	2.0

[015] This example shows that marks made by the pencil lead can be erased more easily if the lead contains hexagonal boron nitride. The other important properties of the pencil lead were not degraded significantly by the use of the hexagonal boron nitride.

5

[016] **Example 2**

Further pencil leads were made according to the method of example 1 and the effect of the lubricant was investigated. The leads tested had the following composition:

- 10 aluminium oxide 35% + hexagonal boron nitride 30% (sum = 65%)
binder: methyl cellulose 7%
dispersant: sorbitan ester 1%
lubricant: as indicated in the table, 17%
pigment: Green No. 7 10%

15

The following results were obtained:

Lubricant	Writing properties	Eraser test	Percentage of rejects
stearic acid	good	4	1
stearic acid 10% and carnuba wax 7%	good	4	2.5
calcium stearate	good	5	1.5
calcium stearate 10% and carnuba wax 7%	good	4	3

[017] **Example 3**

- 5 In a further set of experiments the effect of using different binders was investigated. The methods used to make and test the pencils were the same as in example 1. The lead composition was as follows:

aluminium oxide 35% + hexagonal boron nitride 30% (sum = 65%).

- 10 binder: as indicated in the table below, 7%

dispersant: sorbitan ester 1%

lubricant: stearic acid 17%

pigment: Green No. 7 10%

The following results were obtained:

Binder	Writing properties	Eraser test	Percentage of rejects
Methyl cellulose	good	4	1
hydroxypropyl cellulose	good	4	1.2
Cellink 34b*	good	5	1.2
hydroxypropyl ethyl cellulose	good	4	1

* Cellink 34b is a commercially available hydroxypropyl cellulose binder containing
5 0.5% benzaldehyde, which crosslinks the binder during the drying.

[018] Examples 1-3 have been repeated using mica instead of hexagonal boron nitride
and identical results were obtained. The examples demonstrate that pencil leads which
leave erasable marks can be obtained from a wide range of pencil lead compositions if
10 hexagonal boron nitride or mica is used.

Claims

1. Pencil lead composition comprising a binder, a dispersant, a pigment and a lubricant characterised in that it contains at least 20% by weight of hexagonal boron nitride or mica.
2. Pencil lead composition according to claim 1 characterised in that it consists of
hexagonal boron nitride or mica: 25-45%
metal oxide up to 60%
binder: 5-10%
dispersant: 0.5-2.0%
lubricant: 10-25%
pigment: 0.2-30%
up to 5% optional components,
with the proviso that the sum of the metal oxide, hexagonal boron nitride and mica contents is 40-85%.
3. A method for making a pencil lead with a composition according to claim 1 comprising the steps of kneading together all of the ingredients with a minimum of water to make a paste, extruding the paste to make the leads and drying the extruded leads at a temperature of 50-150°C for 1-4 hours.

Annex 2 (Communication)

1. Document 1 (see claims, example and paragraphs [004] and [009]) discloses pencil lead compositions containing a filler, a binder, a lubricant, a dispersant and a pigment as well as a bittering agent. The composition may contain hexagonal boron nitride or mica as fillers. The composition in the example had a mica content of 30% by weight. The lead was made by kneading together all of the ingredients with a minimum of water. The paste produced was then extruded to make the leads. The extruded leads were dried at a temperature of 120-150°C for 1 hour. Document 1 is thus novelty destroying for the subject-matter of claims 1-3 (Articles 52(1), 54(1) and (2) EPC).

2. Document 2 (see the example) discloses a coloured pencil containing a lead comprising a filler, a binder, a lubricant, a dispersant and a pigment. The filler used was mica which made up 65% of the total composition. The lead was made by kneading together all of the ingredients with a minimum of water. The paste produced was then extruded to make the leads. The extruded leads were dried at a temperature of 110-120°C for 1-2 hours. Document D2 is thus also novelty destroying for the subject-matter of claims 1 and 3 (Articles 52(1), 54(1) and (2) EPC).

3. The subject-matter of claim 2 is unclear (Article 84 EPC). The sum of the components of the lead composition add up to more than 100%, when the sum of the metal oxide, hexagonal boron nitride and mica contents has its maximum value of 85 wt% (the minimum contents of binder 5%, lubricant 10%, dispersant 0.5% and pigment 0.2% mean that this composition must contain a minimum of 100.7 % of components). A composition whose components must add up to more than 100% is unclear.

4. If the applicant wishes to maintain the application, new claims should be filed which take the above objections into account. Care should be taken to ensure that the new claims comply with the requirements of the EPC in respect of clarity, novelty, inventive step, and if necessary unity (Articles 54, 56, 82 and 84 EPC). Care should further be taken that any amendments do not introduce subject-matter which extends beyond the content of the application as originally filed (Article 123(2) EPC).

5. In the letter of reply, the difference between the new claims and the prior art disclosed in documents 1 and 2 should be indicated. The technical problem underlying the invention in view of the closest prior art and the solution to this problem should be readily derivable from the statement of the applicant (Rule 42(1)c EPC and EPO Guidelines, C-IV, 11.7).

6. In order to facilitate the examination as to whether the new claims contain subject-matter which extend beyond the content of the application as filed, the applicant is requested to indicate precisely where in the application documents any amendments proposed find a basis (Article 123(2) EPC).

Annex 3 (Document 1)

Child-Friendly Coloured Pencils

5 [001] The invention relates to child-friendly coloured pencils.

[002] Pencils are made according to a method which has remained unchanged for many years. Cedar wood is cut into thin slats and grooves are then formed in the slats. Leads are then laid in the grooves and fixed in place with an adhesive. A further matching slat
10 is placed on top of the slat with the leads to make a sandwich and glued in place. Individual pencils are then cut from the sandwich, painted and sharpened. Leads for coloured pencils are made by blending a filler, a binder, a dispersant, a lubricant and a pigment. These materials are kneaded together, extruded and then dried to form the lead.

15

[003] The present inventors have for many years supplied pencils for small children. These pencils may not contain toxic ingredients, as children will often put pencils in their mouths. Fragments of the pencils, if chewed off, can pose a choking hazard. Pencils can temporary stain the child's mouth or be ingested. Although the ingredients of the pencil
20 are harmless, many parents can become very worried if they see their child with a stained mouth or eating pencils. The present invention addresses the problem of children putting pencils in their mouths. The solution to the problem is to add a bittering agent to the pencil lead. The bitter taste will ensure that the child quickly stops chewing the pencil.

25

[004] The filler used may be selected from hexagonal boron nitride, aluminium oxide, titanium dioxide or mica or mixtures of these fillers. The filler helps to determine the writing properties of the pencil. Pencils, with leads containing about 30 weight % of hexagonal boron nitride or mica, are believed to produce erasable marks on paper.
30 This is thought to be due to the layered structure of these fillers.

[005] The binder used is preferably a cellulose derivative or a modified starch, more preferably methyl cellulose. Alternatively a polyacrylate may be used. The dispersant is used to help mix the components of the lead intimately and is preferably a sorbitan ester.

5

[006] The lubricant ensures that the pencil writes smoothly and is typically a blend of a wax with either stearic acid or a metal stearate.

[007] Suitable non-toxic pigments are widely available in a full range of colours.

10 The bittering agent is preferably Angostura bitters, hops or quinine.

[008] A typical composition for a pencil lead in accordance with the invention is as follows:

15 filler: 40-70%

binder: 5-10%

dispersant: 0.5-2.0%

lubricant: 10-40%

pigment: 0.2-30%

20 bittering agent: 0.1-0.5%

All percentages are by weight.

[009] The pencil leads are made by kneading together all of the ingredients with a minimum of water. The paste produced is then extruded to form the leads. The extruded
25 leads are dried at a temperature of 120-150°C for 1 hour and, once they have cooled are then ready to be incorporated into pencils using a standard process. The final shape of the pencil may be freely selected. The pencil for example can have a circular, hexagonal or triangular cross-section. It is also possible to coat the pencil with plastic dots to improve the grip of the pencil.

[010] **Example**

A green pencil lead was prepared by kneading together 30% aluminium oxide, 30% mica, 8% methyl cellulose, 1% sorbitan ester, 10% carnuba wax, 9.7 % stearic acid, 11 % green pigment (alpha green available from Farblos AG) and 0.3 % Angostura
5 bitters. The resulting paste was extruded into a lead and dried at 130°C for 1 hour. The lead was formed into a pencil. It was confirmed by testing that the pencil has an unpleasant taste.

Claims

- 10 1. Coloured pencil comprising a lead made from a filler, a binder, a lubricant, a dispersant and a pigment characterised in that the lead also contains a bittering agent.
2. Pencil according to claim 1 where the bittering agent is selected from hops,
15 Angostura bitters and quinine.

Annex 4 (Document 2)

Coloured Pencils

5 [001] Coloured pencils are well known. The first documented coloured pencil dates from 1823, but they are believed to have been made long before then. Methods for making coloured pencils and lead compositions suitable for such pencils are well known. The conventional method for making coloured pencils is the following:

10 [002] A mixture consisting of a filler, a binder, a dispersant, a lubricant and a pigment is kneaded together with a minimum amount of water to form a paste. The paste is then extruded to form a lead. The lead is then dried. The dried leads are then cut to the desired length, glued into a groove in a wooden slat, and assembled into a sandwich structure with a further slat. Pencils are then shaped from the sandwich.

15

[003] One of the most cost-intensive steps in the process is the drying of the lead. Conventional processes require that the lead is dried for 6 hours at 50°C. This slow drying prevents the lead from warping or breaking during the drying step, but the long times involved slow down the production process and require that large drying ovens are available. The applicant proposes a lead composition which enables the leads to be
20 dried at 110-120°C within 2 hours. The binder used is a cross-linked methyl cellulose. This binder is optionally mixed with polyvinyl alcohol in a weight ratio of 4:1 to 10:1. The cross-linking agent used is typically benzaldehyde. The filler used is mica and is present at 50-70 wt% of the composition. The remaining components are conventional.

[004] **Example**

A lead composition was prepared consisting of:

- mica 65wt.% (filler)
- 5 methyl cellulose 5.8 wt.%, polyvinylalcohol 1wt.% and as a cross-linking agent
- benzaldehyde 0.2 wt.% (binder)
- Dispatrol 0.4 wt.% (dispersant)
- calcium Stearate 18 wt.% (lubricant)
- Igalit Red 10.5 wt.% (pigment)
- 10 Angostura bitters 0.1 wt.% (bittering agent)

The composition was kneaded to a paste with water, extruded to make a lead and dried at 120°C for 1.5 hours. The warping of the lead and breakages led to 1.5% of the leads being rejected. This percentage of rejects is far lower than the acceptable value of 3%.

15

Claim

- 1. Coloured pencil containing a lead comprising a filler, a binder, a dispersant, a lubricant and a pigment, characterised in that the binder is a cross-linked methyl cellulose optionally mixed with polyvinyl alcohol in a weight ratio of 4:1 to 10:1, and that the filler is mica and makes up 50-70 wt% of the composition.

20

Annex 5 (Letter from the applicant)

Urgent by Fax

5 Dear Sir,

Please file a response to the official action as soon as possible. We are very close to agreement with a potential licensee who wishes to take up a very lucrative licence on the technology covered by the patent application. In order to be able to finalise the
10 contracts we need the European Patent Office to agree a set of claims upon which a patent can be granted. We have studied the documents cited by the examiner carefully and do not believe that they should prevent us obtaining a patent as they are not primarily concerned with erasable coloured pencils.

15 You may make the amendments you deem necessary to the claims on file. Please do not amend the description just yet as we wish to avoid the patent application proceeding to grant before the licence is signed. The licensee wishes to make pencil leads with the following composition:

20 Titanium dioxide 30-40% + hexagonal boron nitride or mica 20-30%
Binder: methyl cellulose 6.7% + benzaldehyde (cross linking agent) 0.3%
Dispersant: sorbitan ester 1%
Lubricant: stearic acid 7% and carnuba wax 5%
Pigments: 10-30%

25

Yours sincerely

Mr T.A. Colour

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Claims of Annex 1 (Patent application)

1. Pencil lead composition comprising a binder, a dispersant, a pigment and a lubricant characterised in that it contains at least 20% by weight of hexagonal boron nitride or mica.

2. Pencil lead composition according to claim 1 characterised in that it consists of

hexagonal boron nitride or mica: 25-45%
metal oxide up to 60%
binder: 5-10%
dispersant: 0.5-2.0%
lubricant: 10-25%
pigment: 0.2-30%
up to 5% optional components,
with the proviso that the sum of the metal oxide, hexagonal boron nitride and mica contents is 40-85%.

3. A method for making a pencil lead with a composition according to claim 1 comprising the steps of kneading together all of the ingredients with a minimum of water to make a paste, extruding the paste to make the leads and drying the extruded leads at a temperature of 50-150°C for 1-4 hours.