

EUROPEAN QUALIFYING EXAMINATION 1995

PAPER B CHEMISTRY

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INSTRUCTIONS TO CANDIDATES

In this paper, you should assume that a European patent application for all the Contracting States comprising the appended documents* has been filed and that the European Patent Office has issued the annexed official communication. The paper may include a client's letter containing instructions about the way your client wishes to prosecute the European patent application.

You should accept the facts given in the paper and base your answers upon such facts. Whether and to what extent these facts are used is your responsibility.

You should not use any special knowledge you may have of the subject-matter of the invention, but are to assume that the prior art given is in fact exhaustive.

Your task is now to draft a full response to the official communication. The response should be a letter to the EPO, accompanied, if appropriate, by an amended set of claims. No amendments to the description should, however, be made.

The claims should afford the broadest protection possible while meeting the requirements of the Convention. In your letter of response you should set out your arguments in support of the patentability of the independent claim(s).

If you consider that any part of the application ought to be made the subject of one or more divisional applications, you should in a note, clearly identify the subject-matter of the independent claim of such divisional application(s) and the justification for this. However, it is not necessary to draft the wording of the independent claim for the or each divisional application.

In addition to your chosen solution, you may – but this is not mandatory – give, in a note, the reasons for your choice of solution, for example, why you selected a particular form of claim, a particular feature for an independent claim, a particular piece of prior art as starting point or why you rejected or preferred some piece of prior art. Any such note should however be brief.

It is assumed that you have studied the examination paper in the language in which you have given your answer. If this is not so, please indicate on the front page of your answer in which language you have studied the examination paper. This always applies to candidates who – after having filed such a request when enrolling for the examination – give their answer in a language other than German, English or French.

* These documents do not necessarily constitute the only or best solution to the task set in Paper A.

Description of the Application

The present invention concerns special lead-antimony alloys, their manufacture and processing, and their use. More precisely, it is the result of metallurgical investigations mainly in the field of chemical plant construction, in particular with a view to resolving corrosion problems in urea production plants. These problems should be solved in such a way that the urea produced is also not contaminated by corrosion products.

Numerous solutions have been proposed for reducing corrosion in such plants but these have often been either very costly or very complicated and thus difficult to put into practice, or both.

Changing the chemical reaction as such by adjusting the reaction parameters did not lead to a practicable solution. On the contrary, any reduction in corrosion achieved in this way had the undesirable side-effect of reducing yield to a level which was uneconomical. The search for a solution therefore had to be approached differently, i.e. by selecting the right material for the reactor, with due consideration for the price.

It has been known since the early days of chemical plant construction that lead (Pb) is highly resistant to corrosion from many materials. It is sufficient to mention the lead chamber process for the manufacture of sulphuric acid as well as the use of lead pipes for drinking-water supply. However, lead has been eliminated from the present considerations because of its inadequate mechanical strength. There is thus no need to go into the other disadvantages of lead, such as its poisonous properties. It is also known that lead alloyed even with small quantities of other metals, such as antimony (Sb), tin (Sn) and/or arsenic (As), assumes markedly different properties.

In document I the use of lead alloys was proposed with a view to improving the strength of lead pipes and reducing corrosion. The lead content of these alloys is at least 80% by weight and they contain at least one alloy metal, selected from the group Bi, Sb and Sn in quantities of not more than 15% by weight in each case. Examples of such alloys are Pb/Bi, Pb/Sb, Pb/Sn and Pb/Sb/Sn. The alloys can be produced by melting the metals together and casting in moulds. When solidified, the alloys can be further processed in the traditional manner.

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Document II relates to specially manufactured Pb-Sb alloys with an Sb content of 1 to 8% by weight which are said to have an improved resistance to acids and for which the ability to withstand a pressure in the range 0.15 to 1.0 MPa is reported.

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However, Pb-Sb alloys characterised by very high resistance to corrosion from aggressive media do not yet meet all the requirements for chemical plant construction purposes. More particularly, such alloys still have to meet certain mechanical requirements, such as higher pressures. The problem was therefore to improve the mechanical properties whilst maintaining high resistance to corrosion.

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A way has now been found of manufacturing Pb-Sb alloys with very high chemical resistance and very good mechanical properties. These meet the requirements and the material has the potential for a wide variety of uses in chemical plant construction. If the inside reactor walls, which are in contact with the reaction mixture, are manufactured from this material, because of their

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greater strength a simpler outer construction can be used for the reactor than was hitherto necessary.

The subject-matter of the invention is a lead alloy obtainable by
5 the method described below which contains 1 to 15% by weight of antimony, based on the total weight, and roled articles manufactured therefrom. The invention also relates to their manufacture and technical uses.

10 The processing of lead and antimony is not entirely straightforward even in the binary system, despite the apparent simplicity of composition. Whilst molten Pb and Sb can be mixed in almost any proportions, inhomogeneities can occur in the solid state when they cool, as a result of separation. The properties of the Pb-Sb
15 alloys thus greatly depend on the conditions of manufacture. This also applies near the eutectic, which is known to be 11.1% by weight of Sb. A construction material for chemical production plant must of course be homogeneous if it is to be of good strength, to ensure that within the material there are no variations in its properties and, as a result, minimal corrosion, and
20 that it will provide a high degree of safety and have a long useful life (service life).

In order to achieve this, the alloy must be prepared by a special
25 process.

The alloy is prepared according to the process defined in claim 1.

The process according to the invention is now described on the
30 basis of the binary alloys of Pb and Sb.

For this purpose, Pb and Sb are melted together in suitable quantities. More precisely, the appropriate quantity of lead is usually melted in an induction furnace in inert crucibles and covered with a protective anti-oxidant melt consisting of a salt
5 with a low melting point. The requisite quantity of Sb is added through this protective melt with a strong intermixing action at temperatures normally above 400°C. A proportion of 15% by weight of Sb should on no account be exceeded. The hardness of this material becomes greater, the higher the proportion of Sb in this
10 range, however, at the same time the material becomes more brittle. Above the 15% limit the material is also more likely to contain the inhomogeneities mentioned earlier, which make the product unsuitable for the present purpose. Material with less than 1% by weight of Sb does not have the desired properties.

15 For casting, the protective melt is skimmed off and the molten alloy can then be cast at 400°C in steel moulds for example. Up to this point, our process is no different to that used hitherto in the manufacture of these or similar lead alloys.

20 In the traditional method, preheated moulds are used and the alloys are then allowed to cool in them to ambient temperature. The alloys solidify at above approximately 250°C. They are then processed as desired, for example by rolling or recasting. As
25 already pointed out, further processing in this way would not result in a material corresponding to the present requirements.

For that reason, the still liquid alloy in the moulds is treated differently in the process according to the invention. It is
30 quenched with a cooling agent, immediately after casting, to a temperature of less than 200°C, so that no inhomogeneities can occur.

To reach the strength values required for plant construction, the cast bodies (bars) obtained as described above are heated, before further processing, to a temperature of from 135° to 175°C if they have cooled below that level. They are then rolled within this
5 temperature range, for which purpose a lubricant (usually kerosene) is normally used. The thickness of the cast bodies is reduced with each pass by 10 to 20%. The bars or plates rolled in this way are then finish-rolled with their thickness being reduced again by at least 10%, at a rate of 1 to 5% per pass. The rolling
10 temperature in the finish-rolling operation ranges from room temperature (20°C) to 125°C, depending on the exact composition and the exact requirements to be met by the alloy.

Experiments have shown that the alloys for plant construction,
15 especially in the case of urea production plants, should preferably not contain more than 5% by weight of Sb. Above that limit, the number of cracks which can appear in the finish-rolling operation (as a result of the increased brittleness mentioned) rises steeply and weakens the material. It can also be observed that the mechan-
20 ical strength of the alloy is greater the nearer the finish-rolling temperature is to the upper limit of the stated range. The reduction in thickness after each pass at the finish-rolling stage also influences the mechanical properties. It has also been discovered that those of our alloys which have not undergone the
25 special rolling process were comparatively "softer", i.e. they were not as strong, even though their chemical resistance was the same as that of the finish-rolled product.

This presents the advantageous possibility of constructing a reactor with inner walls, which can come into contact with the reaction mixture, consisting of plates made of the hardened alloy, and of using unhardened alloy of the same composition for the joints between the plates. The chief advantage of such a construction is avoidance of contact corrosion. Another advantage is the simpler structure from the point of view of expansion and contraction as temperatures change: The softer material in the joints between the plates is more ductile and can prevent leakages. When the joint is being cast, steps must be taken, if appropriate by means of additional cooling, to ensure that heat is quickly dissipated so as to avoid inhomogeneities in the joint filling such as those steps already mentioned in connection with the mould casting.

The interaction between the alloy and the urea, and especially the absorption of alloy constituents by the urea, has also been investigated. Urea can be converted into biuret which may be used as an additive in animal feed. It is thus desirable that additional purification of the organic product should not be necessary. No traces of the alloy constituents could be found in the urea manufactured in a plant with reactor walls made from the alloy according to the invention, even when it was left in the reactor for long periods in reaction conditions going well beyond the usual period of contact.

One point which has not yet been discussed is the purity of the metals used in the alloy. The metals would generally be of standard quality, but they should preferably not contain a total of more than 0.05% by weight of impurities as otherwise their properties could be subject to unforeseeable changes which would often

make them unsuitable for the intended purposes. Apart from such impurities, the alloys according to the invention comprise the constituents indicated, which together must add up to 100%.

5 Whilst binary alloys produce the desired results in chemical reactors, the presence of a third alloy metal where the ternary alloy is to be used as a corrosion-resistant slide bearing metal, for example as an axle-bearing metal for moderate loads and speeds, might be desired. Suitable compositions for this purpose
10 have been found to be 84 to 89% by weight of Pb, 6 to 12% by weight of Sb and 2 to 8% by weight of Sn. The alloy metals were melted together as described above. The cast bodies are prepared from these alloys by casting and were hardened by quenching them to the solid state immediately after casting in the way described
15 above, and cooled further directly by means of the cooling agent.

The following examples are intended to further illustrate the invention. All the percentages in compositions relate to the total weight of the alloy.

20

The alloys used in the following examples were in all cases manufactured in the same way by melting the alloy components together as described above, and the resulting cast bodies were manufactured by casting in moulds and quenching to below 200°C.

25

Example 1

In this example, not only was the influence of the composition
30 investigated, but also the influence of the temperature and of the reduction in thickness after each pass in the finish-rolling process on the properties of alloys according to the invention. The results are summarised in Tables 1 and 2.

Table 1

Influence of Sb content and the temperature at which finish-rolling was carried out with 1% reduction in thickness after each of 12 passes

5

	Temperature	Sb	Tensile strength
	(°C)	(% by weight)	(MPa)
10	20	1.0	20.3
	20	2.0	22.4
	20	5.0	25.2
	20	10.0	25.2
	125	1.0	23.1
15	125	2.0	28.0
	125	5.0	34.6
	125	10.0	34.7
	130	2.0	19.5
	160	2.0	18.7
20	160	5.0	18.5
	200	2.0	17.8
	225	10.0	13.7

The results show that the tensile strength increases with the Sb content but that as soon as the Sb content reaches 5%, added Sb has scarcely any further influence and some small cracks appear. Finish-rolling at a higher temperature with a fixed Sb content results in higher strength, but above 125°C the effect is reversed and tensile strength falls below the minimum value acceptable for plant construction, namely 20 MPa.

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Table 2

Influence of reduced thickness per pass in the finish-rolling process with an Sb content of 5% by weight

5	Temperature (°C)	Thickness reduction (%)	Tensile strength (MPa)	Surface
	125	1	34.6	(a)
	125	3	34.4	(a)
10	125	5	33.8	(b)

(a) completely smooth (b) hair cracks in places

Example 2

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This example is designed to investigate the influence of the alloys on the purity of urea manufactured in reactors made out of different alloys.

20 Urea was manufactured in accordance with the usual well-known continuous process from NH_3 and CO_2 via the ammonium carbamate intermediate stage. During the process, pressure of 10 to 30 MPa builds up in the reactor at 160 to 200°C.

25 In an experimental reactor I, three experimental plates were used as substitutes for existing plates. All the plates were the same size. Plate 1 was of the standard type used in state-of-the-art plant and was made of pure nickel (Ni), plate 2 was made of a lead-bismuth (Pb-Bi) alloy with 2.5% of Bi and plate 3 consisted
 30 of a Pb-Sb alloy with 2.5% of Sb which had, in accordance with the invention, been quenched, rolled twice at 175°C (with 15% thickness reduction at each pass) and finish-rolled six times at

110°C with 2% thickness reduction per pass. After two months of continuous operation, the reactor was switched off and the experimental plates were removed. The plates were given a visual inspection and the corrosion depth was measured (see Table 3).

5

Table 3

10	Plate 1	entire surface attacked, average depth 3 mm
	2	partially attacked (approx. 25% of surface), average depth 1.5 mm
	3	a few small marks (approx. 1 mm in diameter), 0.1 mm deep.

15 The positive effect of the Pb-Sb alloy is clearly demonstrated. However, the slight traces of corrosion on plates 3 were not evenly distributed, but were concentrated on the areas of the plates which had been in contact with the plates of other materials. Some Ni and traces of Pb and Bi were found in the urea.

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In an experimental reactor II of the same type, in which all the plates were of the above plate 3 type, the plate joints were also cast with an alloy of the same composition as the plates. The plates dissipate so much of the heat that this rapid cooling prevents inhomogeneities from being formed in the joints. In the urea produced in this reactor, there were found no traces of either lead or antimony. The plates showed even less corrosion than in the reactor I described above.

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Example 3

In order to verify resistance to corrosion, test strips from plates of the composition given in table 4 were immersed in fuming sulphuric acid at 45°C for six weeks. The test strips were then removed, rinsed carefully and dried. Resistance to corrosion was measured on the basis of weight loss. A Pb-Bi alloy with 3% Bi (the material normally used in the lead chamber process) was used for comparison.

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Table 4

Each Pb-Sb plate was triple-rolled at 150°C with 10% thickness reduction after each pass, finish-rolled at 125°C and with 1% thickness reduction after each of 12 passes.

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Pb-Bi		10.0% loss
Pb-	0.1% Sb	8.0%
	0.5%	6.0%
	1.0%	0.2%
	1.5%	0.09%
	2.5%	0.06%
	3.0%	0.08%
	5.0%	0.1%
	6.0%	4.0%

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Although the alloy with 6% Sb is still highly corrosion-resistant, the material is already mechanically less suitable for heavy-duty industrial use: the corrosion advances along cracks in the material. No such cracks were observed in alloys with a lower Sb content.

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Claims

1. Process for the manufacture of a lead alloy containing 1 to 15% by weight, based on the total weight of the alloy, of antimony, in which a molten homogeneous mixture of the alloy metals is quenched to a temperature of less than 200°C directly after casting.
2. Process for the manufacture of roled articles from the alloy prepared in accordance with claim 1, characterised in that, after quenching, the alloy is rolled at 135 to 175°C, with the thickness being reduced after each pass by 10 to 20%, after which it is finish-rolled at 20 to 125°C, the total thickness being reduced by at least 10% and by 1 to 5% after each pass.
3. Pb-Sb alloys obtainable by the process according to claim 1.
4. Rolled articles from Pb-Sb alloys having increased tensile strength of at least 20 MPa, obtainable by the process according to claim 2.
5. Rolled articles according to claim 4 which, apart from impurities, consist exclusively of Pb and Sb.
6. Rolled articles according to claim 5, containing not more than 5% by weight of Sb.
7. Use of rolled articles according to any of Claims 4 to 6 for the preparation of reactor walls for chemical production plants.
8. Use of the rolled articles according to any of claims 4 to 6 as a construction material resistant to corrosive media.
9. Use of an alloy according to claim 3 as a slide bearing metal, the alloy apart from impurities, consisting of 84 to 89% by weight of Pb, 6 to 12% by weight of Sb and 2 to 8% by weight of Sn.

Communication

1. Lead alloys containing antimony and/or tin in the concentrations according to the claimed alloys are already known from documents I and II mentioned in the application. The manufacture of such alloys is also described for example in documents I, II and III. Furthermore, the advantageous properties of such alloys, i.e. hardness, tensile strength and resistance to corrosion (e.g. from acids), are mentioned in the documents.
2. Document III refers to the need for the alloys to be pure. It also addresses the problems associated with separation of such alloys and proposes that these difficulties be overcome by quenching the material to below 200°C directly after casting. It also describes in detail the subsequent processing by rolling and finish-rolling in the claimed temperature ranges and with the features mentioned in the claims.
3. In view of these facts, the process according to claims 1 and 2 lacks novelty (Arts. 54(1) and (2) EPC).
4. Document III addresses the fact that both hardness and brittleness increase with the proportion of antimony in the alloy. It discusses this in connection with Sb concentrations of 1 to 5% by weight, the range preferred according to the present claim 6. As the alloy compositions and the steps in the process for increasing hardness and tensile strength are the same, the alloys and the rolled articles according to claims 3 to 6 likewise cannot be considered to be novel.

5. Document IV describes the manufacture of urea and expressly recommends the use of reactors made of rolled Pb-Sb-alloy plates. Document III recommends improvements to the properties of such alloys. For this reason, claim 7 clearly does not involve an inventive step.
6. Claim 8 is likewise not allowable owing to lack of an inventive step, because the term "corrosive media", in our view, comprises everything which causes corrosion, and documents I and III disclose alloys according to the claimed composition which feature increased resistance to corrosion. Document II refers in connection with such alloys to their resistance to acids.
7. You should, however, be aware that, since claims 1 to 6 are not allowable for the reasons given, if they are deleted, claims 7 to 9 can no longer be regarded as possessing unity in accordance with Article 82 EPC.
8. You are invited to submit claims which take the aforementioned objections into account and which comply with the requirements of the EPC, especially with regard to novelty, inventive step, clarity, disclosure in the documents as filed and unity (Arts. 54(1) and (2), 56, 84, 123(2) and 82 EPC).

In your reply you should also identify the difference between the new claims and the state of the art and its significance,

and present the invention in such a way that both the technical problem to be solved vis-à-vis the state of the art and the solution found (see Rule 27(1)(c) EPC), and your position on the matter of inventive step can be understood.

According to the Guidelines (C-III, 4.4) an independent claim should specify clearly all of the essential features needed to define the invention, i.e. each independent claim must indicate all the features which are necessary to solve the problem on which the invention is based.

9. Attention is drawn to the fact that the application may not be amended in such a way that its subject-matter extends beyond the content of the application as filed (Art. 123(2) EPC). Therefore, and also in view of Guidelines E-II, 1 and C-VI, 5.4, it should be explained from where in the original application documents new features in newly formulated claims have been directly and unambiguously derived.

DOCUMENT I (State of the Art)

(The document is identical to document I in paper A)

Water pipes have for a long time been made from lead. Manufacturing such pipes is a very simple and reliable process. The pipes can be easily adapted to fit the requirements of the building site due to their good bending qualities and are well-suited for soldering, which is also an advantage later in the event of repairs. Furthermore, one of the reasons for using lead pipes for water supply is that a coating of low solubility forms on the surfaces which protects the pipe against corrosion.

10 However, the known pipes have a number of serious disadvantages. Lead is very soft, and the mechanical strength of the pipes is therefore inadequate over the long term. This disadvantage is particularly serious at threaded connections subject to mechanical loads. The coating is repeatedly damaged at points where the lead
15 pipe is subject to mechanical loads. Drinking water often contains foreign ions which attack the coating of the lead pipes.

The use of pure lead pipes in water pipe construction is thus no longer tolerable in the long term.

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In contrast to this situation, we have now found that various lead alloys have greatly improved properties and are not only suitable for water pipe construction but, because of their improved hardness, tensile strength and resistance to corrosion are also suitable
25 for other uses. To cite just a few examples, they can be used

for printers' type, bearing metal, roofing or as a repair material for other metallic materials. The alloys according to the invention are very suitable for soldering and can themselves be used as soldering material.

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The subject-matter of the invention is lead alloys having a lead content of at least 80% by weight and with at least one alloy metal selected from the group bismuth, antimony and tin in quantities of 1 to 15% by weight in each case. Other elements, such as
10 poisonous arsenic, should be kept below 0.3% by weight.

Good examples of our alloys are (percentages by weight):

Pb 95/Bi 5; Pb 89/Sb 11; Pb 97.5/Sb 2.5; Pb 97.5/Bi 2.5;

Pb 97.5/Sn 2.5; Pb 89/Sb 7/Sn 4; Pb 86/Sb 9/Sn 5.

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The alloys can be obtained by simply melting the metals together, by dissolving the added metals in molten lead or by mixing the molten metals at a temperature of about 400°C. To prevent oxidation it has often proved advantageous to cover the surface of the
20 molten metals with a protective melt of salts having a low melting point, which can be easily removed before solidification. The molten alloys are cast in moulds and left to solidify. They can then be processed further in the traditional manner to form pipes or plates.

DOCUMENT II (State of the Art)

(The document is identical to document II in paper A)

The use of acids requires materials resistant to these products. Experiments have shown that lead-antimony alloys in a suitable composition manufactured by a special method, give satisfactory results. This new alloy can replace the traditional lead or enamel
5 lining. Manufacture is quite simple: lead is melted in a hermetically sealed furnace; antimony is melted nearly in another container. When the antimony has turned dark red, it is poured into the lead and thoroughly stirred and the alloy is then allowed to cool.

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The table gives the pressure which the alloy is able to withstand at a given wall thickness (% = % by weight)

	Pressure in MPa	% Sb	Thickness in cm.
15	0.15	1.0	3.0
	0.2	2.0	3.0
	0.2	3.0	2.0
	0.3	4.0	2.0
	0.5	5.0	2.5
20	0.6	6.0	3.0
	0.8	7.0	3.5
	1.0	8.0	4.0

DOCUMENT III (State of the Art)

LEAD-ANTIMONY ALLOYS

1. Production of lead-antimony alloys

The corrosion-resistant alloys are manufactured by stirring antimony below its melting point into molten lead or, conversely, by adding lead to molten antimony. The alloy constituents can also be melted separately and the melts can then be mixed together.

The Pb-Sb phase diagram was recently revised on the basis of a critical review of publications. The solidifying temperature of the eutectic can accordingly be taken as $252 \pm 0.5^{\circ}\text{C}$ and its composition, as 11.1% by weight of Sb.

As lead is capable of absorbing not more than 3.45% by weight of Sb in solid solution, alloys with a lower Sb content should not contain a eutectic. However, even in cases with substantially lower percentages of Sb and in the normal cooling process, the precipitation of separate crystals of lead, antimony and eutectic may be observed. Even in the case of alloys with only 0.1% by weight of Sb this tendency is very marked, indicating that these alloys have a heterogeneous structure.

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Where cooling of the alloys is normal or delayed, the two alloy constituents, antimony and lead, separate. Both hardness and tensile strength drop sharply as a result. Such separation may be counteracted by, for example, casting the alloy in a cold mould and, better still, by quenching the cast material

immediately after casting. In this process it is essential that the material should solidify quickly, so that the inhomogeneities mentioned are prevented from forming. They can be reliably prevented by quenching to below approximately 200°C.

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2. Mechanical properties and processing of the materials

The strength properties of lead-antimony alloys, especially in the range of 1 to 5% by weight of Sb, depend to a very great extent on the previous history of the material and its homogeneity. Depending on the kind of pre-treatment, strength values in the stated concentration range can differ by more than 100%. As with tensile strength, hardness also depends on the previous treatment. Even small additions of other alloy constituents and impurities can have a major influence on the strength properties of the material. Quantities of such other constituents and impurities from 0.05% by weight, based on the total weight of the alloy, may alter and even impair these properties - tensile strength, for example.

As is clear from the foregoing, the hardness and strength of Pb-Sb alloys increases substantially if they are quenched after casting.

These properties can be further improved (tensile strengths in excess of 30 MPa) in the case of a given composition of the alloy if the cast bodies are subjected to a subsequent mechanical working stage. An example of suitable subsequent working is, in particular, rolling at an elevated temperature, but well below the softening temperature, i.e. in the range up to

170 or 180°C at most. This process is usually carried out with a thickness reduction rate of between 15 and 20% per pass of the roller.

5 Further improvements in these properties can be attained by means of additional cold- or finish-rolling, i.e. at temperatures around or slightly below 125°C. In these processes the thickness of the material is again reduced, usually by
10 10 to 20%. However, this must take place in smaller stages in order to obtain smooth surfaces. Generally, thickness is reduced by approximately 1 to 5% per pass. It should be noted that subsequent mechanical working becomes more difficult the higher the proportion of Sb, as this also increases hardness and brittleness. For many applications this results in a
15 natural limit slightly above the eutectic. Tensile strengths of up to 35 MPa can thus be attained and high pressures can also be withstood.

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DOCUMENT IV (State of the Art)

Urea is one of the basic compounds used in a number of chemical reactions. Suffice it to mention polymer chemistry and its "duro-plastics" or the thermal polymerisation of the urea, with ammonia being separated off to biuret, triuret etc. A frequent requirement
5 is that the urea should, as far as possible, contain no impurities. There have accordingly been various recommendations concerning the use of corrosion-resistant reactor material. In connection with such citations from the literature, lead and its alloys with tin or antimony, glass or nickel and its copper alloys
10 may be mentioned. Metals are generally preferred on account of their better mechanical properties - i.e. tensile strength and resistance to wear.

Liquid ammonia and gaseous carbon dioxide can be converted under
15 pressure and at an elevated temperature to ammonium carbamate. This compound can be decomposed by heat to urea and water. To achieve a high degree of conversion to urea, a marked excess of ammonia is generally used, as in this way, the balance of the overall reaction can be shifted to the right:

20



A solution of ammonium carbamate, liquid ammonia and carbon dioxide was converted in a reactor. The reactor was made of rolled
25 plates of a homogeneous lead-antimony alloy, the joints between the plates of which were filled with pure lead. The reactants were continuously fed in a mole proportion of ammonia to carbon dioxide of 2.5 - 3 at a pressure of 250 to 300 bar (25 - 30 MPa) and converted in the reactor at 190 - 200°C. Because of the high
30 ammonia content, a conversion rate, based on CO₂, of over 70% was achieved. By lowering the pressure on the reaction mixture after leaving the reactor to 60 - 80 bar in a high-pressure separator, the greater part of the excess ammonia and the water evaporates. By lowering the pressure still further and applying heat, the
35 remaining carbamate is again decomposed into the starting substances and separated from the urea. The recovered ammonia and carbon dioxide are recycled.

Client's letter

Dear Sir/Madam,

In view of the patent office's comments concerning our patent application, which we have now received, we wish to inform you that we are no longer interested in pursuing the application insofar as the use of the ternary Pb-Sb-Sn alloy as a bearing metal is concerned, because further investigations have revealed a number of disadvantages with regard to its wearing properties, and the material would therefore no longer appear to be suitable for the use envisaged.

Now that the new production plant which our client has ordered from us is about to be built, we request you to do your utmost to obtain the broadest possible protection, at least for our plants and in particular for the reactor described in the application.

For the rest, we rely on your professional judgment as to what extent a successful case can be made for more far-reaching protection and request you to take whatever steps may be necessary to that end.

Yours faithfully,

Reinvent GmbH & Co. KG
Production Plant
Builders