

INTERNATIONAL SMELTING AND REFINING COMPANY

25 Broadway

New York

OFFICE OF
THE VICE PRESIDENT

January 30, 1942

Mr. E. O. Sowerwine, Secretary,
International Smelting & Refining Co.,
B u i l d i n g.

Re: Johnson et al. Application, Serial
No. 178,352, For: ELECTROCHEMICAL
METHOD OF PRODUCING WHITE LEAD

Dear Sir:

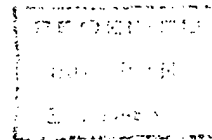
I am sending you herewith for filing, the
original deed in the above mentioned application,
which is assigned to the International Smelting &
Refining Company,

Will you kindly acknowledge receipt of
this deed on the attached carbon copy and return
for my files.

Yours very truly,

Frederick Laist

KS/
Encl.



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D4947

January 29, 1942

1-20-42

Mr. Frederick Laist, Vice-President
International Smelting and Refining Co.
25 Broadway (Room 1720)
New York, N. Y.

Dear Sir: Re: Johnson et al. Application, Serial No.
178,352, For: Electrochemical Method of
Producing White Lead - Now patent No.
2,270,783

Enclosed herewith is the original deed in the
above-identified application, together with five printed
copies for your files. This deed is an original document
of title and should be carefully preserved.

It will be appreciated if you will acknowledge
safe receipt of this document.

Very truly yours,

James W. Laist

Copy of letter and 5 copies of patent sent Mr. Case

PNYC00008395

N11206.01

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January 2, 1942

1-3-42

Mr. Frederick Laist, Vice-President
International Smelting and Refining Co.
25 Broadway (Room 1720)
New York, N. Y.

Dear Sir: Re: Johnson et al. application, Serial No.
178,352, Filed December 6, 1937, For:
Electrochemical Method of Producing
White Lead

We have paid the final fee in the above-mentioned
application, and have been advised that the patent will
issue January 20, 1942, as patent No. 2,270,783.

Very truly yours,

James W. Laist

Copy of this letter sent Mr. F. O. Case - 1/3/42

PNYC00008396

N11206.02

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June 30, 1941
ACKNOWLEDGED RECEIPT

Mr. Frederick Laist, Vice-President
International Smelting and Refining Company
25 Broadway (Room 1720)
New York, N. Y.

Dear Sir: Re: Johnson et al. Application, Serial
No. 178,352, Filed December 6, 1937,
For: Electrochemical Method of Pro-
ducing White Lead

This is to advise you that we have received a
notice of allowance in the above-mentioned application.
The final fee of \$30.00 is due on or before December 28,
1941, and unless you instruct us to the contrary, we
will defer payment of this fee until shortly before the
due date.

Very truly yours,

James W. Laist

cc: Mr. F.O. Case

PNYC00008397

N11206.03

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D4947

June 27, 1941

ACKNOWLEDGED RECEIPT

6-30-41

Mr. Frederick Laist, Vice-President
International Smelting and Refining Co.
25 Broadway (Room 1720)
New York, N. Y.

Dear Sir: Re: Johnson et al Application, Ser. No.
178,352, Filed December 6, 1937

We have just received notice that the Examiner has changed the title of the above-mentioned application from "Method of Producing White Lead" to "Electrochemical Method of Producing White Lead". It is not usual for the Examiner to make such a change, except when the application is in condition for allowance, and consequently I anticipate receipt of notice of allowance in the near future.

Very truly yours,

Sam W. Laist

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KENNETH H. MURRAY
CARL E. RING
W. B. MORTON, JR.

June 23, 1941

ACKNOWLEDGED RECEIPT

6-24-41

Mr. Frederick Laist, Vice-President
International Smelting and Refining Company
25 Broadway (Room 1720)
New York, N. Y.

Dear Sir:

Re: Johnson et al. Application, Ser. No.
178,352, Filed December 6, 1937,
For: Method of Producing White Lead

After filing our appeal in the above-mentioned application, the Examiner apparently changed his mind about the allowability of the claims, and called our Washington representative to state that, with certain minor amendments, he felt that the claims could be allowed. We have prepared a Supplemental amendment for the purpose of making the changes suggested by the Examiner, and we understand that this amendment will probably result in allowance of the case without the necessity of going through with the appeal. Two copies of the Supplemental amendment by which these changes were made are enclosed herewith.

Very truly yours,

Sam W. Laist

Enc.

Copy of letter with enclosure sent Mr. Case

PNYC00008399

N11206.05

MADE IN U.S.A.

IN THE UNITED STATES PATENT OFFICE

In re application of :

CUNHARD E. JOHNSON, :
REGINALD G. BOWMAN and :
WILLIAM J. MOX :
BY :

Serial No. 178,352 :

METHOD OF PRODUCING
WHITE LEAD

Filed Dec. 6, 1937 :

Div. 56 - Room 4725 :

New York, N.Y., June 21, 1941.

Honorable Commissioner of Patents

Washington, D.C.

SUPPLEMENTAL AMENDMENT

S i r :

In further response to the Status Letter of
January 28, 1941, please amend the above-identified applica-
tion as follows:

In the Specification:

Page 6, line 8, after the period insert --"Excess
carbonation", as used herein and as
understood in the art, means carbonation in
excess of that required to maintain the
bicarbonate ion concentration of the anolyte
at the value necessary for the production
of a white lead product of desired
composition.--

Page 6, line 8, after the period insert --"Each of
these procedures effects the desired con-
trol of the bicarbonate ion concentration
in the anolyte, and hence the composition

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N11206.06

Excerpt

of the precipitated white lead, in a manner such that the addition of ions not entering into the composition of the precipitated white lead is not substantially greater than the loss of such ions in the course of normal operation of the cell.--

In the Claims:

Cancel claims 2, 4, 5 and 11.

Add the following claims:

-- 23. In a process of producing white lead of constant particular composition by electrolysis in a bifluid cell involving the use of an anolyte capable under electrolysis of yielding a lead solvent and containing a bicarbonate and of a catholyte containing a carbonate which is altered during the course of the process by removal of carbonate ions and involving carbonation of the catholyte to restore carbonate ions thereto and re-use of the regenerated catholyte, the composition of the precipitated white lead being controlled by reference to the bicarbonate ion concentration in the anolyte, the improvement which comprises adjusting the bicarbonate ion concentration in the anolyte by subjecting the catholyte to excess carbonation and by incorporating hydroxyl ions in the anolyte in the form of a soluble metal hydroxide in such an amount that the amount of added metal ions of said hydroxide is not substantially in excess of the amount of metal ions lost in the course of normal operation of the cell, the amount of metal hydroxide incorporated in the anolyte being sufficient to establish therein bicarbonate ions in the desired concentration, whereby the addition of ions not

entering into the composition of the precipitated white lead is not substantially greater than the loss of such ions in the course of normal operation of the cell.

24. In a process of producing white lead of constant particular composition by electrolysis in a bifluid cell involving the use of an anolyte capable under electrolysis of yielding a lead solvent and containing a bicarbonate and a catholyte containing a carbonate which is altered during the course of the process by removal of carbonate ions, and involving carbonation of the catholyte to restore carbonate ions thereto and re-use of the regenerated catholyte, the composition of the precipitated white lead being controlled by reference to the bicarbonate ion concentration in the anolyte, the improvement which comprises adjusting the bicarbonate ion concentration in the anolyte by subjecting the catholyte to excess carbonation and adding to the anolyte a sufficient amount of uncarbonated catholyte containing hydroxyl ions to establish in the anolyte bicarbonate ions in the desired concentration, whereby addition of ions not entering into the composition of the precipitated white lead is not substantially greater than the loss of such ions in the course of normal operation of the cell.

25. In the process of producing white lead of constant particular composition by electrolysis in a bifluid cell in which the white lead is precipitated in an anolyte capable under electrolysis of yielding a

lead solvent and containing bicarbonate ions. The steps which consist in determining the bicarbonate ion concentration in the anolyte corresponding to the precipitation of white lead of the particular composition, and maintaining said bicarbonate ion content in the anolyte by maintaining a uniform and excess carbonation of the catholyte and by adding an alkali metal hydroxide to the anolyte in such an amount that the amount of added alkali metal ions is not substantially in excess of the amount of alkali metal ions lost in the course of normal operation of the cell but sufficient to establish in the anolyte bicarbonate ions in the desired concentration for precipitation of white lead of the particular composition.

27. In a process of producing white lead of constant particular composition by electrolysis in a bi-fluid cell involving the use of (1) an anolyte capable under electrolysis of yielding a lead solvent and containing a carbonate and a bicarbonate and (2) a catholyte containing a carbonate which is altered during the course of the process by removal of carbonate ions, and involving carbonation of the catholyte to restore carbonate ions thereto and re-use of the regenerated catholyte, the composition of the precipitated white lead being controlled by reference to the bicarbonate ion concentration in the anolyte, the improvement which comprises maintaining the carbonate concentration in the anolyte at or below a predetermined maximum which is substantially less

than the concentration resulting in objectionable deposition of white lead on the anode, and adjusting the bicarbonate ion concentration in the anolyte by subjecting a portion of the catholyte to excess carbonation and by adding to the anolyte the remaining portion of uncarbonated catholyte containing sodium hydroxide to establish in the anolyte bicarbonate ions in the desired concentration, whereby the concentration of ions not entering into the composition of the precipitated white lead is not increased in the course of normal operation of the cell. --

R E M A R K S

Reconsideration of this application in its amended form is respectfully requested.

Applicants' solicitor wishes to acknowledge an interview courteously granted by the Examiners in charge of this application. At the interview new claims 23 to 27 were considered as to matters of form and as to patentability. It is understood that the new claims in their present form are considered allowable.

New claims 23 to 27 are similar to former claims 2, 4, 5 and 11. The new claims differ from the former claims in matters of form and the manner in which the amount of added hydroxide ions is defined.

The specification has been amended on page 2 to point out, in conformity with the language of the claims, each of the procedures for controlling the carbonation

PNYC0008404

concentration in accordance with the invention effects the desired control of the bicarbonate ion concentration, and hence the composition of the precipitated white lead, in a manner such that the addition of ions not entering into the the composition of the precipitated white lead is not substantially greater than the loss of such ions in the course of normal operation of the cell. In this connection it is noted that the Status Letter contains the statement that: "Inspection of the affidavit indicates that the building up of the sodium ion concentration is detrimental (last paragraph of page 2), yet claim 4 recites the addition of a sodium salt". The affidavit points out that in procedure (4) of the Johnson et al patent the amount of sodium bicarbonate which must be added to effect control of the anolyte bicarbonate ion concentration is so great that sodium ions are built up in the electrolyte at a rate greater than the rate at which the sodium ions are lost in the course of normal operation of the cell. It is true that in the control method of the present invention sodium ions, in the form of sodium hydroxide, are added to the anolyte, but the amount of hydroxide which must be added in accordance with this invention to effect the desired control is so much less than in procedure (4) of the Johnson et al patent that the required amount of added sodium hydroxide does not add sodium ions to the electrolyte any faster than these sodium ions are lost in the course of normal operation of the cell.

In view of the fact that there appears to be some confusion as to the meaning of "excess carbonation" the following discussion is offered by way of explanation. White lead is a product comprising the chemical combination of lead ions, carbonate ions and hydroxyl ions. The quantitative

ratio of carbonate to hydroxyl radicals in the white lead product determines the physical characteristics of the product. A substantial range of variation is possible. If we assume that a white lead product of constant particular composition is to be produced in a bifluoride cell, it is apparent that a substantially constant ratio of concentration of carbonate ions to hydroxyl ions must be maintained in the anolyte from which the white lead product is precipitated. Carbonate ions and hydroxyl ions present in the anolyte are used up as the white lead product is precipitated. Bicarbonate ions are formed as a by-product of the electrochemical action taking place within the cell (see page 4, lines 15 and 16, of the specification). Hydroxyl ions, formed electrolytically at the cathode, build up a relatively high concentration of hydroxyl ions in the catholyte.

In order to replenish the supply of carbonate ions in the anolyte for precipitation of the white lead product of particular composition, it is conventional practice to add carbon dioxide to the catholyte where it reacts with hydroxide to form the corresponding carbonate. Thus, as the degree of carbonation of the catholyte proceeds the amount of hydroxyl ions present in the catholyte decreases and carbonate ion concentration increases. The porous diaphragm separating the anolyte and catholyte permits passage of the carbonate and hydroxyl ions from the catholyte (where their concentration is relatively high) into the anolyte. By adjusting the degree of carbonation of the catholyte it is possible to establish in the catholyte a ratio of concentration

of carbonate and hydroxyl ions such that these ions pass through the diaphragm into the anolyte in exactly the quantitative ratio that these ions combine in the formation of the desired white lead product. However, as pointed out above, bicarbonate ions are produced in the anolyte during the course of electrolysis. Bicarbonate ions and hydroxyl ions are mutually incompatible in the same solution; a high bicarbonate ion concentration holds down the hydroxyl ion concentration and vice versa. Thus, the high concentration of bicarbonate ions produced in the anolyte tends to reduce the effective concentration of hydroxyl ions introduced into the anolyte from the catholyte. Conversely, an increase in the hydroxyl ion concentration in the anolyte will decrease the bicarbonate ion concentration in the anolyte by changing the bicarbonate to carbonate. Summarizing, therefore, an equilibrium exists in the catholyte between carbonate and hydroxyl ions, and the ratio of concentrations of these ions may be controlled by the degree of carbonation of the catholyte. An equilibrium also exists in the anolyte between hydroxyl and bicarbonate ions. It is possible to select a degree of carbonation of the catholyte such that the concentration of hydroxyl ions in catholyte (and passing into the anolyte) is high enough to reduce the bicarbonate ion concentration in the anolyte to the point where the desired hydroxyl ion concentration, in proportion to the carbonate ion concentration, can exist in the anolyte. The amount of carbonation of the catholyte required to accomplish this result is known as complete carbonation. The patent to Harrington

represents an operation wherein this "complete" carbonation is effected. It is difficult to carbonate completely because complete carbonation requires quantitatively accurate addition of CO_2 to the anolyte. Addition of any amount more than the quantitatively exact amount of CO_2 to the catholyte for complete carbonation is "excess carbonation", and additions of any amount less than such quantitatively exact amount of CO_2 is "deficient carbonation". The specification has been amended on page 6 to incorporate therein a definite statement of what is meant by "excess carbonation".

In the Johnson et al patent the catholyte is subjected to "deficient" carbonation. As a result, the ratio of hydroxyl ions to carbonate ions in the carbonated catholyte is relatively high, and the resulting migration of carbonate and hydroxyl ions from the deficiently carbonated catholyte into the anolyte is more than enough to reduce the bicarbonate ion concentration in the anolyte to the point where the desired hydroxyl ion concentration can be maintained. In order to overcome this undesirably high concentration of hydroxyl ions in the anolyte a bicarbonate or an acid is added to the anolyte, and control of the white lead product composition is thereby obtained.

In the present application the catholyte is subjected to excess carbonation, that is, an amount of carbonation in excess of that required to maintain the bicarbonate concentration of the anolyte (and hence the hydroxyl ion concentration of the anolyte) at the value desired for the production of a white lead product of constant particular composition. Excess carbonation, as will be understood from

the foregoing discussion, establishes in the anolyte an amount of carbonate (with respect to the hydroxyl ion concentration) greater than that required for complete reaction with the material with which the carbonate reacts to produce the white lead product of particular composition. This definition of excess carbonation was presented in the amendment dated October 13, 1939. It will be seen, therefore, that the second and third sentences in the third paragraph of the amendment dated October 13, 1939 are not inconsistent. The bicarbonate ion concentration in the anolyte is an indicator of the concentration of hydroxyl ions in the anolyte. In accordance with the present invention, the bicarbonate ion concentration in the anolyte is adjusted by incorporating hydroxyl ions in the anolyte in an amount sufficient to establish therein such an amount of bicarbonate ions that it is possible for carbonate and hydroxyl ions migrating from the catholyte to accumulate in the anolyte in the desired concentration ratio, and so to produce white lead of particular uniform composition. This method of controlling the composition of the precipitated white lead is different from the methods described in the Harrington and the Johnson et al patents.

It is believed, accordingly, that the claims of the present application are now allowable in their present form and that the application is in condition to be passed to issue.

Favorable reconsideration and early allowance are respectfully requested.

Respectfully submitted,

Attorneys for Applicants