

气相过氧化氢的生成及大气中浓度的测定*

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摘要 研究了 HC-NO₂-空气体系中 H₂O₂ 和有机氢过氧化物的生成。模拟实验结果表明: H₂O₂ 生成浓度的最大值与碳氢化合物的初始浓度成正比; 生成规律与 O₃ 生成规律类似, 比值 [O₃]_{max}/[H₂O₂]_{max} 随 [HC]₀ 增大而减小。在乙烯、戊烷体系中, H₂O₂ 在总过氧化物中的比例为 0.6 左右; 而丙烯体系中, 随光照时间的延长, 有较多的有机氢过氧化物产生。大气监测结果显示: H₂O₂ 的生成受污染物排放、日照及温度的影响, 早晚浓度低, 午后形成峰值, 日变化规律与 O₃ 浓度变化相似, 监测到的最大浓度值在春季为 1.3×10^{-9} , 在秋季为 0.64×10^{-9} 。

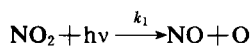
关键词 过氧化氢, 模拟实验, 大气监测。

在光化学污染中产生的二次污染物过氧化氢(H₂O₂)越来越引起人们的重视, 这不仅是由于其本身是氧化剂, 对人体等有刺激作用, 更重要的在于它是一个非常活跃的化学物质, 可分解为 OH 自由基而参加一系列化学反应, 特别是其对大气上层云水中二氧化硫(SO₂)的氧化起着重要作用。这一氧化反应对酸雨的形成是相当重要的。有些学者甚至认为在水汽和雾滴存在的条件下, H₂O₂ 对 SO₂ 的氧化起决定性作用^[1-4]。由此对大气中 H₂O₂ 形成和存在的研究, 也就显得更加重要了。

本文对 HC-NO₂-空气体系光化学反应中 H₂O₂ 的生成规律进行了探讨, 并对北京地区大气中的 H₂O₂ 浓度进行了有目的的观测。

1 实验部分

选择 HC-NO₂-空气体系观测 H₂O₂ 在光化学反应中的生成规律。反应在聚全氟乙丙烯膜烟雾箱中进行^[5]。该箱呈圆柱型, 体积约 1.2m³, 周围配有 160 根 20W 黑光灯管做光源, 并配有净化空气系统。光强反应式:



$$k_1 = 0.127 \pm 0.010/\text{min}$$

每次实验前, 用净化空气洗反应箱 2—3 次, 然后向箱中充净化空气约 120min, 流量 10L/min; 在通气结束前 20min 将 HC 和 NO₂ 注入箱中。通气结束后静置 30min, 以使反应物混合均

匀。开灯照射后, 记录不同时刻的 NO₂、O₃ 和 H₂O₂ 浓度值, 并取样监测 HC 浓度值。

NO₂ 用化学发光式 NO-NO₂-NO_x 分析仪测定, O₃ 用化学发光式臭氧测定仪分析, HC 用 GC-7A 气相色谱仪(氢火焰检定器, β, β-氧二丙腈层析柱)测定。H₂O₂ 和总过氧化物(Total Hydroperoxides, 以下简称 THP, 包括 H₂O₂ 和有机氢过氧化物(Organic Hydroperoxides), 以下简称 OHP), 用美国国家大气研究中心研制的双道 H₂O₂-THP 分析仪测定^[6,7], 可同时测出 H₂O₂ 和 THP 的浓度。

对北京地区大气中 H₂O₂ 浓度的测定选择在不同季节, 并观测了 H₂O₂ 浓度日变化曲线。

2 结果和讨论

2.1 C₂H₄-NO₂-空气体系

图 1 给出了 C₂H₄-NO₂-空气体系在光照过程中物质浓度-时间变化曲线。从图 1 可以看出, H₂O₂ 与 THP 在光照约 150min 时出现一峰值, 随后呈下降趋势, H₂O₂ 与 THP 的变化趋势一致, 由于体系中很难产生大量的有机氢过氧化物, 故在 THP 中 H₂O₂ 所占份额是较大的。H₂O₂ 最大浓度值出现的时间比 O₃ 最大浓度值出现的时间大约晚 60min, 而 NO₂ 在光照 30min 后很快下降至

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一个很低的浓度水平。此体系的实验数据列于表 1。从实验结果看出,在 NO_2 初始浓度基本不变的条件下, C_2H_4 的初始浓度越大, H_2O_2 的浓度最大值也越大;当 C_2H_4 的初始浓度不变时, H_2O_2 浓度最大值随 NO_2 初始浓度值的增大而增大。 O_3 的生成和存在对 H_2O_2 的生成有影响, H_2O_2 最大浓度值与 O_3 最大浓度值成正比关系。

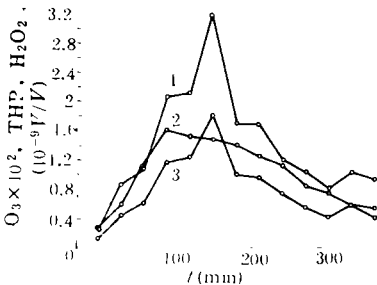


图 1 $\text{C}_2\text{H}_4\text{-NO}_2$ -空气体系生成物浓度随时间变化曲线(反应 3)
1. THP 2. O_3 3. H_2O_2

表 1 HC-NO_2 -空气体系数据(10^{-9}V/V)

HC	$[\text{NO}_2]_0$	$[\text{H}_2\text{O}_2]_{\max}$	$[\text{THP}]_{\max}$	$[\text{O}_3]_{\max}$	$\frac{[\text{O}_3]_{\max}}{[\text{H}_2\text{O}_2]_{\max}}$
$[\text{C}_2\text{H}_4]_0$					
1670	417	0.84	1.53	125	149
3330	417	1.49	2.98	134	90
5000	420	1.87	3.18	160	88
6670	410	2.70	4.53	195	72
8300	422	3.82	6.36	225	59
170	83	0.7	1.08	94	134
172	120	1.54	2.44	102	69
165	167	2.23	3.35	84	37
174	220	2.73	4.62	152	55
167	330	3.02	5.16	192	63
$[\text{C}_3\text{H}_6]_0$					
208	292	1.29	1.46	126	98
408	294	2.34	2.58	209	89
898	290	2.11	2.72	202	95
1800	293	3.09	4.92	238	77
2450	300	7.41	12.33	257	35
3270	329	7.44	12.10	168	23
4080	294	9.41	10.77	105	11
$[\text{C}_5\text{H}_{12}]_0$					
770	71	0.84	1.49	120	142
3550	83	1.70	2.72	122	72
4620	88	1.00	1.76	80	80
16720	88	3.52	5.88	165	47
34600	86	4.01	6.35	142	35

2.2 $\text{C}_3\text{H}_6\text{-NO}_2$ -空气体系

图 2 为 $\text{C}_3\text{H}_6\text{-NO}_2$ -空气体系在光照过程中物种浓度-时间变化曲线。从图 2 可以看到,在光照约 80min 后出现 O_3 浓度最大峰值,随即出现 H_2O_2 和 THP 的最大浓度峰值。而在丙烯体系中, H_2O_2 和 THP 最大浓度峰值的出现要到 150min 左右,这反映出丙烯的光化学反应活性远大于乙烯。特别值得一提的是,在达到 H_2O_2 和 THP 最大值后, H_2O_2 浓度呈逐渐减小的趋势,而 THP 则下降后又有升高,出现第二个峰值,这表明在丙烯体系中,产生了较多的有机氢过氧化物(OHP)。实验数据列于表 1。从实验结果可以看出,在其他条件不变的情况下,随 C_3H_6 初始浓度的增大, H_2O_2 、THP 和 O_3 的最大浓度值增大。

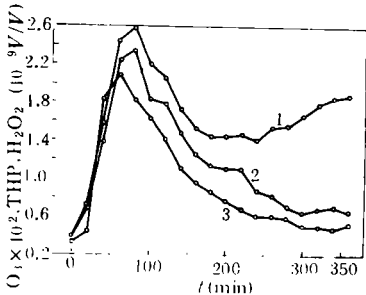


图 2 $\text{C}_3\text{H}_6\text{-NO}_2$ -空气体系生成物浓度随时间变化曲线(反应 2)
1. THP 2. O_3 3. H_2O_2

2.3 不同反应体系之间的比较

在模拟实验中,还观测了 $\text{C}_5\text{H}_{12}\text{-NO}_2$ -空气体系中 H_2O_2 的生成。从图 3 可以看出,由于烷基的反应活性较低,戊烷(C_5H_{12})体系中达到 H_2O_2 、THP 和 O_3 浓度最大值的时间在 400min 左右,比烯烃体系要慢得多。实验结果列见表 1。

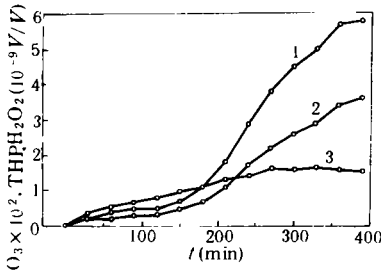


图 3 $\text{C}_5\text{H}_{12}\text{-NO}_2$ -空气体系生成物浓度随时间变化曲线(反应 4)
1. THP 2. O_3 3. H_2O_2

对比3个不同反应体系的观测结果,可以看到:随HC初始浓度的增加, H_2O_2 、THP和 O_3 浓度最大值增加,但3个体系中达到最大值所需的时间不同,丙烯体系所需时间最短约80min,乙烯体系需150min,戊烷体系需400min,这反映了3个体系光化学反应活性的不同。

在3个体系中,生成物 H_2O_2 与 O_3 浓度之间有着密切的联系,一般来说, H_2O_2 最大浓度值增大,则 O_3 最大浓度值也增大。但如果对 $[\text{O}_3]_{\max}/[\text{H}_2\text{O}_2]_{\max}$ 比值进行研究,则发现这个比值随 $[\text{HC}]_0$ 的增大而减小(见表1)。可以预测,在碳氢排放较严重的地区, H_2O_2 对S(N)的氧化作用会变得更加重要。

H_2O_2 和 O_3 作为光化学反应的二次污染物,在生成曲线上有着相似的情况(见图1—3),即 H_2O_2 和 O_3 会同时做为氧化剂存在。虽然 O_3 的浓度比气态 H_2O_2 浓度高2个数量级,但 H_2O_2 的亨利常数比 O_3 的大3个数量级,故水相中对S(N)的氧化仍然是 H_2O_2 占重要地位。

在3个体系中, H_2O_2 浓度与THP浓度间的关系有所不同。在乙烯和戊烷体系,随光照时间的延续, $[\text{H}_2\text{O}_2]/[\text{THP}]$ 之值基本稳定在0.6左右,其表明乙烯和戊烷由于其结构特点,难以生成大量的有机氢过氧化物。而丙烯体系则不同,在反应中, H_2O_2 和THP首先达到一最大值,而后 H_2O_2 不断下降,THP在经过一段时间后又呈上升的趋势,表明随光化学反应的进行,丙烯体系中产生较多的有机氢过氧化物,致使 H_2O_2 在THP中所占比例逐渐减小(图4)。

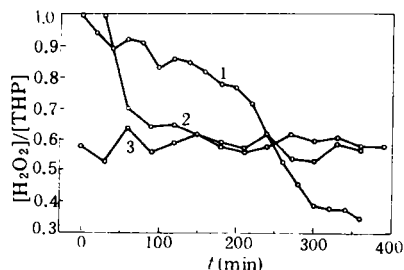


图4 $[\text{H}_2\text{O}_2]/[\text{THP}]$ 随光照时间变化曲线

1. 丙烯体系 2. 戊烷体系 3. 乙烯体系

2.4 北京地区大气中气态 H_2O_2 浓度测定

对大气的实际监测分别于1991年5月3日

至6日,9月11日至19日及10月16日至18日在北京大学校园内昼夜进行。

在测定 H_2O_2 浓度时,同时测定了大气中 O_3 和 NO_2 的浓度。图5给出了 H_2O_2 、 O_3 和 NO_2 浓度的日变化曲线。从图5可看出:在一天中, NO_2 浓度最大值在10:00左右, O_3 和 H_2O_2 浓度最大值在14:00左右。因为在清晨交通量大,汽车排放污染物多,日照又弱,利于 NO_2 积累,故出现 NO_2 峰值;随日照的增强, O_3 和 H_2O_2 浓度提高,峰值出现在午后,且 H_2O_2 与 O_3 浓度密切相关。

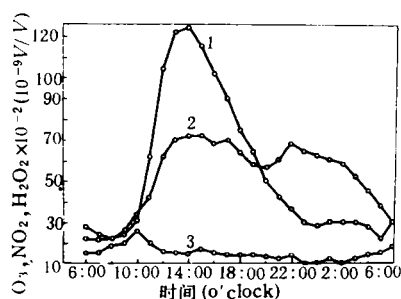


图5 空气中 $[\text{H}_2\text{O}_2]$ 、 $[\text{O}_3]$ 和 $[\text{NO}_2]$ 浓度日变化曲线

(1991年5月5日)

1. H_2O_2 1. O_3 3. NO_2

图6给出了大气中THP、OHP和 H_2O_2 浓度日变化曲线。大气监测数据列于表2。从监测结果可以看出: H_2O_2 浓度受天气变化的影响,晴天日照强,温度高,其浓度就高。在春季 H_2O_2 浓度高于其在秋季的浓度。在一天中, H_2O_2 浓度随日照的增强而提高,早晚浓度低,午后形成峰值,其变化规律与 O_3 浓度变化规律相似。 H_2O_2 比OHP对气候条件的变化更敏感,但二者的变化趋势是相似的。在THP浓度低时,以OHP为主。

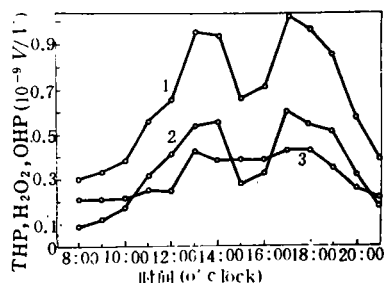


图6 大气中 $[\text{THP}]$ 、 $[\text{H}_2\text{O}_2]$ 和 $[\text{OHP}]$ 日变化曲线

(1991年9月13日)

1. THP 2. H_2O_2 3. OHP

表 2 大气气态 H₂O₂ 观测浓度 (10⁻⁹, I'/I')

日期(月-日)	05-03	05-04	05-05	05-06	09-11	09-12	09-13	09-14	09-17	09-18	09-19	10-16	10-17	10-18
天气	晴	晴间多云	晴	晴	晴	晴	晴	阴	阴	晴	多云转晴	晴	晴	晴
最高温度(℃)	21.4	22.0	23.0	23.0	24	24	25	22	22	21	20	20	14	15
[NO ₂] _{max}	30	30	25	25										
[O ₃] _{max}	60	65	75	70										
[THP] _{max}	0.80	0.89	1.01	0.84	0.62	0.62	0.73	0.63	0.34	0.26				
[H ₂ O ₂] _{max}	0.6	0.9	1.3	1.2	0.64	0.60	0.59	0.50	0.25	0.45	0.50	0.42	0.13	0.09

3 小结

(1)在 3 个 HC-NO₂-空气体系中,均有 H₂O₂ 生成;相比之下,丙烯体系产生 H₂O₂ 浓度高,生成反应迅速。

(2)H₂O₂ 与 O₃ 的生成有相同的趋势。

(3)在乙烯、戊烷体系中,H₂O₂ 在 THP 中所占比例为 0.6 左右,且这比值不受其他因素的影响。丙烯体系中,在达到 H₂O₂ 和 THP 最大浓度值后,H₂O₂ 浓度连续下降,而有机氢过氧化物(OHP)又出现新的峰值。

(4)大气监测的结果表明 H₂O₂ 的生成受日照、温度的影响,在大气中 H₂O₂ 浓度变化规律与 O₃ 的规律相似。

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Abstracts

Chinese Journal of Environmental Science

Semiconductor Photocatalytic Degradation of Anionic Sodium Dodecylbenzenesulphonate in Aqueous Solutions. Jiang Weichuan and Wang Qiquan (Dept. of Chemistry, Zhejiang University, Hangzhou 310027); *Chin. J. Environ. Sci.*, 15(6), 1994, pp. 1—3

Anionic sodium dodecylbenzenesulphonate (DBS) can be photodegraded in aqueous suspension of TiO_2 powder by irradiation with a 300W high pressure mercury lamp. The photocatalytic ability of anatase and rutile TiO_2 in the solutions at different pH values has been studied. The effects of adding H_2O_2 and Cu^{2+} on the photodegradation have also been discussed. The results show that anatase TiO_2 has a more efficiently catalytic ability than rutile TiO_2 . H_2O_2 can reduce the rate of degradation in a basic solution. Cu^{2+} can increase the rate of degradation in an acidic solution.

Key words: semiconductor, photocatalysis, sodium dodecylbenzenesulphonate.

Application of the H_2O_2 - O_3 Process for the Treatment of Wastewaters from the Production Process of Naphthalene-sulfonate and Anthraquinone Sulfonic Acid Dye Intermediates. Yang Zhihua, Zhu Wanpeng et al. (Dept. of Environ. Eng., Tsinghua University, Beijing 100084); *Chin. J. Environ. Sci.*, 15(6), 1994, pp. 4—7

A H_2O_2 - O_3 process in which hydroxyl free radical ($\text{OH}\cdot$) is generated and acts as a non-selectively strong oxidizer was developed and applied to treating refractory wastewaters from the production process of naphthalenesulfonate and anthraquinone sulfonic acid dye intermediates before a biological treatment. It was found that this treatment process could quickly decompose refractory organics in the wastewaters and had a higher ability to improve their biodegradabilities than other conventional methods. Thus treated wastewaters could also be coagulated and flocculated more effectively. It is also found that some groups of the aromatic compounds in the effluents from this treatment process were substituted with one or more OH groups, indicating that the decomposition of organics in the treatment with this newly developed process may follow a hydroxyl free radical mechanism.

Key words: dye intermediates, wastewater treatment, O_3 , H_2O_2 , hydroxyl free radical, biodegradability.

Study on the Microbial Film Immobilization Methods for Preparing the Biosensors for Rapid BOD Determination. Liu Baohong et al. (Dept. of Chemistry, Fudan University, Shanghai 200433); *Chin. J. Environ. Sci.*, 15(6), 1994, pp. 8—11

Several physical and chemical methods, such as the sodium alginate-calcium chloride embedding process, the cellulose acetate sandwiching process and the bovine serum albumen-glutaraldehyde cross-linking process, were used to prepare the immobilized microbial films which were then combined with an oxygen electrode to make the newly developed biosensors for rapid BOD determination. The tests were carried out for the effects of different films on the performances of electrode, including its response sensitivity, linearity ranges, response time, stability and life-time. The performances of the biosensors which were formed under different process conditions such as pH values, temperatures, cross-linking agents and the amounts of embedding agents used were also compared. The results show that the biosensors prepared with the cross-linking process have the most preferred performances, including an electrode life-time of up to 40 days, a BOD determination time of 10 to 15 minutes, and the reproducibility and accuracy of determinations both in compliance with the requirements of standard BOD determination.

Key words: biosensor, biological oxygen demand (BOD), microbial electrode.

Formation of Hydrogen Peroxide in Gaseous Phase and the Determination of Its Atmospheric Concentrations. Zhang Jianbo and Tang Xiaoyan (Center for Environ. Sci., Peking University, Beijing 100871); *Chin. J. Environ. Sci.*, 15(6), 1994, pp. 12—15

Studies on the formations of H_2O_2 and organic hydrogen peroxides in the HC- NO_2 -dry air systems were carried out in an indoor smog chamber. The results from simulated experiments show that the maximum concentrations of H_2O_2 formed are directly proportional to the initial concentrations of hydrocarbons (HC)₀; H_2O_2 is formed by following a similar rule to that of ozone (O_3), and the ratio of (O_3)_{max} / (H_2O_2)_{max} decreases with increasing (HC)₀. In the system of ethylene or pentane, the ratio of H_2O_2 to total hydroperoxides (THP) is about 0.6; and in the system of propylene, more organic hydrogen peroxides are formed with extending irradiation time. The results from atmospheric monitoring show that the formation of H_2O_2 is dependent on emission of pollutants, sunshine and temperature, and it has a lower concentration in both morning and evening and a peak concentration occurred after noon, following a daily change pattern similar to that of ozone. The atmospheric H_2O_2 was found to have a maximum concentration of 1.3×10^{-9} in spring and of 0.64×10^{-9} in autumn as monitored in Beijing in 1991.

Key words: hydrogen peroxide, simulated

Abstracts

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experiment, atmospheric monitoring.

Study on the Biodegradabilities of Dyes under the Aerobic Conditions. An Huren (China- Japan Friendship Environ. Protection Centre, Beijing 100029), Qian Yi et al. (Dept. of Environ. Eng., Tsinghua University, Beijing 100084); *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 16—19

Three simple and practical aerobic biodegradability test methods, i. e., static flask screening test, Warburg respirometry and semi-continuous activated sludge system, were chosen depending on the characteristics of dyes to test the biodegradabilities of 26 water soluble dyes. The results of tests using these three methods were compared and the reasons for the differences between these results were explained. The effects of environmental factors on the biodegradability tests were also discussed. It was found that most of the dyes studied are refractory under the aerobic conditions, and the semi-continuous activated sludge system is a favourable and more precise test method although it takes a longer time.

Key words: dyes, biodegradability, aerobic.

Study on the Mechanism of the Bacteria- added Biological Contact Oxidation Process in the Treatment of Wastewater from Jiemycin Production Process. Luo Guowei and Yang Danqing et al. (Institute of Environ. Sci., South China Normal University, Guangzhou 510631); *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 20—22

This paper deals with a treatment system in which the "hydrolytic acidification - two staged biological contact oxidation-coagulation" process was used to treat a high strength wastewater from the production process of jiemycin. Particularly, it relates to the characteristics, distribution patterns and degradative functions of its aerobic microbial films. The selection and breeding of efficiently degradative bacteria, and whether or not the added bacteria can keep its dominance in the reactor were also studied to explore the mechanism of treating jiemycin wastewater by using the bacteria-added biological contact oxidation process. By the separation and identification of aerobic microbial films from this system, 18 strains bacteria species in 11 genera were obtained. Their distribution of bacteria counts and the effectiveness of degrading organics show that after passing through a pilot operation the added bacteria still exist in the reactor and take the dominant position, and the most dominant strain was identified to be in *Aeromona*. Both qualitative and quantitative analyses of the effluents from pilot aerobic treatments were made to find the reasons for causing the remaining COD_{Cr} values.

Key words: jiemycin wastewater, bacteria- added biological contact oxidation process, dominant species of bacteria, biodegradability, GC/MS.

Classification of Atmospheric Pollution Areas and Afforestation Models. Zhao Yong and Li Shuren (Henan University of Agriculture, Zhengzhou 450002); *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 23—27

Based on the monitoring and predictive analysis of atmospheric pollutants, the clustering analytic method and synthetic index method were used to classify the pollution areas into four types and to classify the greening vegetations into three types in terms of the size of index value. Then further based on the features of pollution for each type of pollution areas, corresponding afforestation models and plant species were selected to green the areas in order to improve the atmospheric environmental quality there. The study demonstrates that more serious single and compounded pollutions generally took place at 300 to 4000m leeward from pollution sources, where an afforestation model consisting of the proper combination of arbor, bush and grass was better in improving the quality of atmospheric environment.

Key words: afforestation model, atmospheric pollution, synthetic index.

Study on the Database System for the Biodegradabilities of Organics in Wastewaters. Huang Xia and Jiang Bin (Dept. of Environ. Eng., Tsinghua University, Beijing 100084); *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 28—32

Based on analyzing in an all-round way the status of demand for the database systems for biodegradabilities of organics in wastewaters and their features, and by using a software engineering method, the systems analysis and systems design of the users- oriented database system have been completed that form a firm basis for further implementing the system.

Key words: wastewater, organics, degradability, database system.

Study on the Electrode Polarization in the Electrolytic Treatment of Lead Wastes Containing Gold and Silver. Zhang Zhongyan, Liang Huqi et al. (Shanghai University of Technology, Shanghai 200072); *Chin. J. Environ. Sci.*, **15**(6), 1994, pp. 33—37

A detailed study, based on the theories of electrochemical thermodynamics kinetics, was carried out on the polarization behaviour and process conditions in the electrode processes of separating gold and silver from lead and obtaining a pure lead