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Kinetics of reduction of lead chloride and oxychloride with sodium carbonate

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Abstract. This study investigates the kinetic behaviour of the reduction reaction between lead chloride ($PbCl_2$) and lead oxychloride ($PbO·PbCl_2$) with sodium carbonate in the presence of carbon in a reducing atmosphere. The relevance of this research stems from the need to develop effective technologies for processing lead-containing secondary production dusts and to optimise the composition of sodium-containing fluxes. Thermodynamic evaluation of the reactions using the Outotec HSC Chemistry software package demonstrated that, within the temperature range of 500-1000°C, both reactions exhibit negative ΔG values and high equilibrium constants, indicating their thermodynamic feasibility. Kinetic studies were carried out using thermogravimetric analysis under isothermal conditions in the temperature range of 800-1100°C for the $PbCl_2$–Na_2CO_3–C system and 500-800°C for the $PbO·PbCl_2$–Na_2CO_3–C system. It was established that the reductive interaction proceeds via a multistage mechanism with a change in the rate-controlling step as the degree of conversion increases. For the $PbO·PbCl_2$–Na_2CO_3–C system, the initial stage of the process is satisfactorily described by the first-order reaction equation, indicating the predominance of chemical control; the apparent activation energy at this stage is 33.5 kJ/mol. As the degree of conversion increases, the activation energy rises to 62-80 kJ/mol, which indicates a transition to a regime complicated by diffusion and structural limitations. The $PbO·PbCl_2$–Na_2CO_3–C system is characterized by higher reactivity and rapid attainment of significant conversion degrees. At moderate conversion levels, the process proceeds predominantly under chemical control ($E_a \approx 43\text{-}44$ kJ/mol); however, at $\alpha \geq 55\%$, an increase in activation energy up to 74 kJ/mol is observed, indicating an increasing influence of mass transfer. It is shown that the presence of oxygen in the structure of lead oxychloride significantly affects the kinetic characteristics of the reductive process in sodium-containing flux systems. The obtained results can be used in optimizing technological schemes for processing lead-containing dusts in order to enhance metal recovery and improve the energy efficiency of the process.

Keywords: *lead-containing dust, lead chloride, lead oxychloride, sodium carbonate, reductive electric smelting, solid-state reaction kinetics, activation energy, thermogravimetric analysis.*

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1. Introduction

Secondary lead accounts for a significant share of the global market, with approximately half of the world's lead consumption being met through the recycling of lead-containing waste [1-4]. According to analytical data [5], the global secondary lead market is estimated at approximately USD 18.7 billion in 2025 and is projected to grow to about USD 26.3 billion by 2035, with an average annual growth rate of around 3.5% (Figure 1). The high recycling rate is driven by both economic and environmental factors, including the relative ease of scrap collection and a reduction in energy consumption by up to 35-40% compared to the production of primary lead from ore [5].

The main contribution to the secondary lead market comes from the recycling of battery production waste, including lead-containing dusts generated during the manufacture and recycling of lead-acid batteries [6-7]. These materials contain lead, antimony, chlorine, and a number of associated elements which, under rational processing conditions, can serve as sources of additional raw materials [8, 9]. However, a significant portion of such fine dust waste accumulates at industrial sites, creating environmental burdens while simultaneously representing an underutilized resource [10]. This highlights the need to develop efficient processing technologies that ensure both minimization of environmental impact and recovery of valuable components.

Dust waste from battery recycling represents complex multicomponent materials. Their composition includes various lead compounds – sulfates ($PbSO_4$), sulfides (PbS), oxides (PbO), oxysulfates ($PbO·PbSO_4$), and chloride compounds ($PbCl_2$, $PbO·PbCl_2$) – as well as impurities of Zn, Sb, Cd, and As [8, 11].

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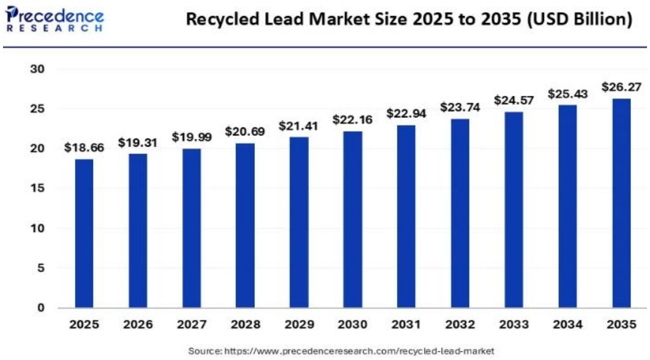

Source: https://www.precedenceresearch.com/recycled-lead-market

Figure 1. Forecast of the secondary lead market growth (2025-2035)

Differences in chemical reactivity and thermochemical behavior of these compounds necessitate a differentiated approach to their processing.

Electrothermal methods, particularly reductive electric smelting using sodium salts, demonstrate significant potential for the comprehensive recovery of lead and associated elements [12-14]. Sodium salts used as fluxes can be partially replaced by calcium compounds (carbonate or oxide), which are capable of binding chlorine from lead chloride compounds and transferring it into the slag phase. This contributes to improved process selectivity, reduced metal losses, and expansion of the raw material base for fluxing agents. In the present work, primary attention is focused on studying the kinetic regularities of the reductive interaction of lead chloride compounds ($PbCl_2$ and $PbO \cdot PbCl_2$) with sodium carbonate in a carbon-containing medium. Lead compounds of other classes, as well as systems involving calcium-containing fluxes ($CaCO_3$, CaO), were not considered in this series of experiments and will be the subject of further investigation.

2. Materials and methods

2.1. Raw materials

Chemically pure substances were used for the kinetic studies to model the main chloride compounds of lead present in battery production dust wastes: lead chloride ($PbCl_2$, analytical grade, ≥99%) and lead oxychloride ($PbO \cdot PbCl_2$, ≥99%). Sodium carbonate (Na_2CO_3, ≥99%) was used as a fluxing additive, and carbon powder (≥99%) served as the reducing agent. The particle size of all components did not exceed 0.074 mm. The sample masses were selected in accordance with the stoichiometry of the reactions under consideration. The use of chemically pure reagents made it possible to eliminate the influence of extraneous impurities and to obtain reproducible data on the kinetic characteristics of the studied systems, thereby providing a basis for the subsequent extrapolation of the results to real industrial materials.

2.2 Experimental procedure

Thermodynamic analysis of the reductive reactions of $PbCl_2$ and $PbO \cdot PbCl_2$ with sodium carbonate in the presence of carbon was carried out using the licensed software package Outotec HSC Chemistry v. 8.1.5 (Reaction Equations module). Calculations were performed in the temperature range of 500-1000°C for stoichiometric reagent compositions with determination of the Gibbs free energy change (ΔG), enthalpy change (ΔH), entropy change (ΔS), and equilibrium constant.

Kinetic studies of the interfacial interaction of lead chloride and oxychloride compounds with fluxing additives and a reducing agent were carried out using thermogravimetric analysis under isothermal conditions. The experiments were performed on a laboratory thermogravimetric setup consisting of a vertical tubular electric furnace with an adjustable working zone, a quartz reaction retort, a continuous mass recording system, and a temperature control unit (Figure 2).

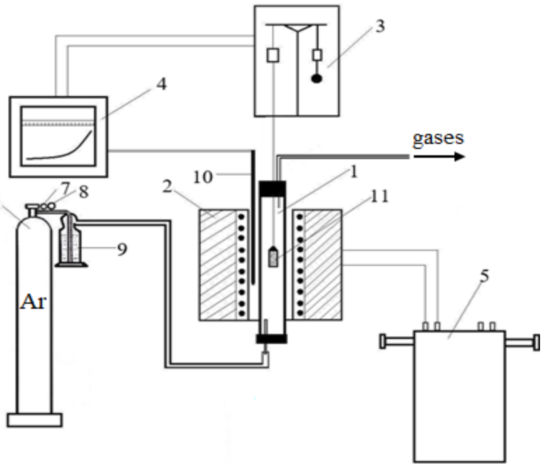

Figure 2. Schematic diagram of the thermogravimetric setup for studying reaction kinetics: 1 – quartz retort; 2 – electric furnace; 3 – ADV-200 analytical balance; 4 – KSP-4 potentiometer; 5 – voltage regulator; 6 – argon cylinder; 7 – rotameter; 8 – manometer; 9 – gas purification system; 10 – tungsten–rhenium thermocouple; 11 – alumina crucible with sample

The samples were heated to the required temperature within the range of 500-1000°C. Isothermal kinetic experiments were conducted primarily in the temperature interval of 800-1100°C, corresponding to the conditions of reductive electric smelting of lead-containing dusts. Temperature control was maintained within ±5°C using a tungsten–rhenium thermocouple and an automatic regulation system. The reactions were carried out in an inert gas atmosphere (argon), supplied to the lower part of the reaction chamber. The argon flow ensured the removal of gaseous reaction products and prevented oxidation of the sample. The sample was placed in an alumina crucible, which was positioned in the isothermal zone of the retort and suspended from analytical balances with a measurement accuracy of 10^{-4} g. The sample was introduced into the working zone after stabilization of the furnace temperature; the time required to reach isothermal conditions did not exceed 1-5 s. The mass change of the sample was recorded continuously throughout the experiment.

Kinetic parameters were determined from the dependencies of the degree of conversion on time at fixed temperatures. The approach to the analysis of kinetic data and the interpretation of temperature dependences was based on classical principles of extractive metallurgy kinetics [15]. The degree of conversion was calculated from the change in sample mass associated with the release of gaseous reaction products. Experimental data were processed using the Arrhenius equation to determine the apparent activation energy and the pre-exponential factor. All experiments were performed in at least three replicates; the deviation of results did not exceed 5%. After completion of the experiment, the samples were cooled to room temperature in an argon atmosphere, followed by control weighing and phase analysis of the reaction products.

3. Results and discussion

3.1. Thermodynamic assessment of the reductive reactions of $PbCl_2$ and $PbO·PbCl_2$ with Na_2CO_3 in a reducing tmosphere

To investigate the reductive interaction of lead chloride and lead oxychloride with sodium carbonate in a reducing atmosphere, two model systems were examined. System I represent the interaction of $PbCl_2$ with sodium carbonate and carbon, while system II represents the reduction of $PbO·PbCl_2$ in the presence of sodium carbonate.

For convenience, these systems are hereafter referred to as systems (1) and (2). The overall reactions are expressed as follows:

$$PbCl_2+Na_2CO_3+C\rightarrow Pb+2NaCl+CO_2(g)+CO(g); \quad (1)$$

$$PbO·PbCl_2+Na_2CO_3+C\rightarrow 2Pb+2NaCl+2CO_2(g); \quad (2)$$

To preliminarily evaluate the thermodynamic feasibility of the reactions, the Gibbs free energy change (ΔG) and equilibrium constant (K) were calculated over the temperature range of 500-1000°C. It was found that, for both reactions, the ΔG values remain negative over the entire investigated temperature range and become more negative with increasing temperature (Table 1). Specifically, for reaction (I), ΔG decreases from −97.9 kJ/mol at 500°C to −244.3 kJ/mol at 1000°C, while for reaction (II) it decreases from −171.5 to −321.0 kJ/mol.

Table 1. Values of the Gibbs free energy change (ΔG) and the logarithm of the equilibrium constant (log K) in the temperature range of 500-1000°C

T, °C	500	600	700	800	900	1000
			Reaction I			
ΔG, kJ	-97.947	-127.217	-155.821	-183.780	-214.968	-244.298
Log K	6.618	7.611	8.365	8.946	9.572	10.024
			Reaction II			
ΔG, kJ	-171.472	-202.021	-231.836	-260.936	-292.879	-320.974
Log K	11.586	12.087	12.445	12.702	13.042	13.170

The log K values for reaction (I) lie within 6.6-10.0, while for reaction (II) they fall within 11.6-13.2, reflecting a substantial thermodynamic driving force and a pronounced shift of equilibrium toward the reaction products. The more negative ΔG values and higher equilibrium constants associated with reaction (II) indicate enhanced thermodynamic stability of the products and agree with the experimentally observed higher reactivity of the $PbO·PbCl_2$-Na_2CO_3-C system.

Therefore, both reactions are thermodynamically feasible throughout the studied temperature range, and the observed differences in reaction rate and changes in the kinetic regime are mainly attributable to kinetic factors rather than thermodynamic constraints.

3.2. Kinetics of the reductive interaction of $PbCl_2$ and $PbO·PbCl_2$ with sodium carbonate in a reducing atmosphere

Although both reactions are thermodynamically feasible throughout the studied temperature range, they exhibit markedly different kinetic behaviors, requiring a detailed examination of the reaction mechanism and rate-controlling stages.

3.2.1. Kinetics of the interaction of $PbCl_2$ with Na_2CO_3 and C (system I)

Analysis of the kinetic data obtained under isothermal conditions in the temperature range of 800-1100°C showed that the interaction of lead chloride with sodium carbonate and carbon proceeds with a change in the kinetic regime as the reaction progresses. At the initial stage of the reaction, at degrees of conversion $\alpha \leq 0.2$, the experimental dependences are satisfactorily described by the first-order reaction equation, indicating the predominance of chemical control. As the interaction time and the degree of conversion increase, deviations from first-order kinetics are observed, associated with a change in the rate-limiting step. At higher degrees of conversion, the process shifts to a transitional regime, indicating a more complex interaction mechanism and the involvement of mass transfer factors and structural changes in the solid phase, which are characteristic of topochemical reactions in the solid state [16].

Figure 3 presents the dependences of the degree of conversion on time for system I at various temperatures. An increase in temperature leads to process acceleration and the attainment of high conversion degrees within a shorter time, which served as the basis for the subsequent calculation of reaction rates and kinetic parameters.

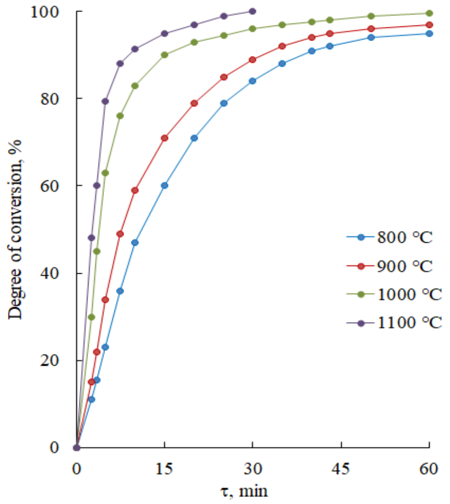

Figure 3. Dependence of the degree of conversion on time for system I ($PbCl_2$–Na_2CO_3–C)

Based on the experimental $\alpha(\tau)$ dependences, the reaction rate was calculated as $v = d\alpha/d\tau$. The dependences of the reaction rate on the degree of conversion for system I are presented in Figure 4.

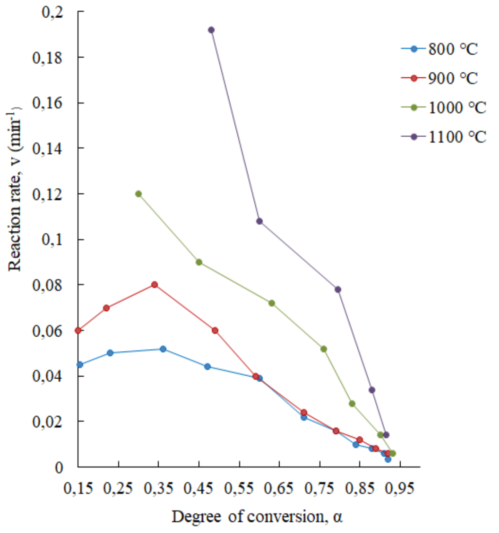

Figure 4. Dependence of the reaction rate on the degree of conversion (α) for system I ($PbCl_2$–Na_2CO_3–C)

Analysis of the obtained data shows that, at identical degrees of conversion, the reaction rate increases with increasing temperature, particularly at the early stages of the process ($\alpha \approx 0.2$-0.4). As the degree of conversion increases, a decrease in the reaction rate and a convergence of the curves obtained at different temperatures are observed, indicating a change in the rate-limiting step and a transition from chemical control to a mixed kinetic regime. At 900°C, a pronounced maximum is observed in the rate versus conversion dependence in the region of $\alpha \approx 0.3$-0.35, which is associated with the intensification of the chemically controlled stage at the initial stage of interaction. At a lower temperature (800°C), this effect is less pronounced due to the slower activation of the reaction, whereas at higher temperatures (1000-1100°C), the rate maximum shifts toward lower degrees of conversion and is partially not detected experimentally because of the high reaction rate.

For the initial stage of the process, described by the first-order reaction equation ($\alpha \approx 0.2$), the apparent activation energy determined from the analysis of the reaction rate constants (K) is 33.5 kJ/mol. The temperature coefficient of the rate constant varies only slightly and lies within the range of 1.4-1.7. The obtained value reflects the kinetics of the chemically controlled stage and corresponds to an idealized description of the initial reaction period.

To evaluate the temperature dependence of the initial reaction rate, the dependence of ln v_0 on $10^3/T$ was analyzed, where v_0 is the reaction rate at the first recorded time point ($\tau = 2.5$ min). The resulting relationship (Figure 5) is linear ($R^2 \approx 0.97$), and the calculated apparent activation energy is about 62 kJ/mol. The higher E_a value compared to that of the chemically controlled stage is attributed to experimental factors and the onset of structural and diffusion limitations at elevated temperatures.

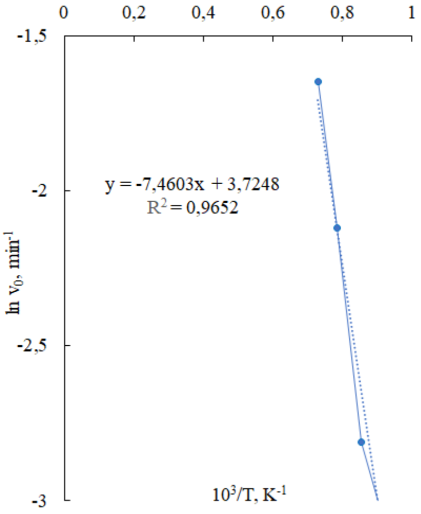

Figure 5. Dependence of ln v_0 on $10^3/T$ for system I ($PbCl_2$–Na_2CO_3–C)

In the region of higher degrees of conversion for system I, temperature dependences of ln k versus $10^3/T$ were constructed (Figure 6). The linear character of the obtained relationship ($R^2 \approx 0.98$) confirms the applicability of the Arrhenius equation. The calculated apparent activation energy is approximately 80 kJ/mol, indicating a significant complication of the interaction mechanism and a transition from chemical control to a regime influenced by mass transfer and the formation of a dense product layer.

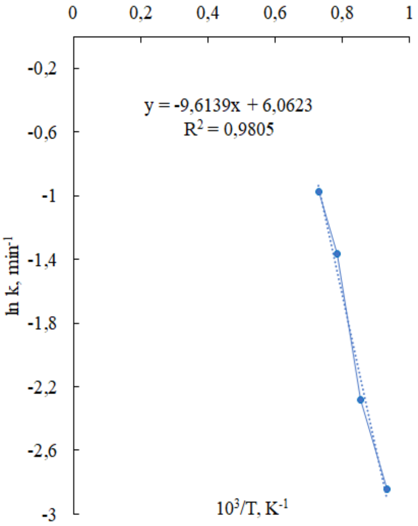

Figure 6. Dependence of ln k on $10^3/T$ for system I ($PbCl_2$–Na_2CO_3–C)

The reductive interaction of $PbCl_2$ with Na_2CO_3 and C proceeds via a multistage mechanism with a change in the kinetic regime as the reaction progresses. At the initial stage, the reaction follows first-order kinetics and is characterized by a relatively low apparent activation energy (33.5 kJ/mol), indicating the predominance of chemical control. As the degree of conversion and temperature increase, the influence of structural and diffusion factors becomes more pronounced, leading to an increase in the apparent activation energy to approximately 62-80 kJ/mol. The obtained results indicate a transition from a chemically controlled stage to a regime influenced by mass transfer and the formation of a dense product layer [16].

3.2.2. Kinetics of the interaction of $PbO \cdot PbCl_2$ with Na_2CO_3 and C (system II).

The kinetics of the reductive interaction of lead oxychloride with sodium carbonate and carbon were investigated under isothermal conditions in the temperature range of 500-800°C. In contrast to system I, this system is characterized by high reactivity, manifested in the rapid attainment of significant degrees of conversion already at the initial stages of the process. Similar acceleration of reductive processes in the presence of sodium fluxes has previously been reported during the processing of lead-containing materials and concentrates [13, 14].

Figure 7 presents the dependences of the degree of conversion on time for system II at various temperatures. An increase in temperature leads to a sharp acceleration of the process: at 700-800°C, the degree of conversion reaches approximately 90-95% within 30-40 min, whereas at 500-600°C it amounts to about 56-65% over the same period. The shape of the curves is characterized by an intensive increase in conversion at the initial stage followed by a gradual slowdown of the process, indicating a change in the rate-limiting step as reaction products accumulate and the structure of the solid phase evolves. Due to the high reaction rate and the rapid attainment of significant degrees of conversion, the identification of a linear region corresponding to first-order kinetics and the accurate determination of the rate constant k for system II are complicated. Therefore, the temperature dependence of the kinetic parameters was analyzed based on the apparent activation energy values determined at fixed degrees of conversion.

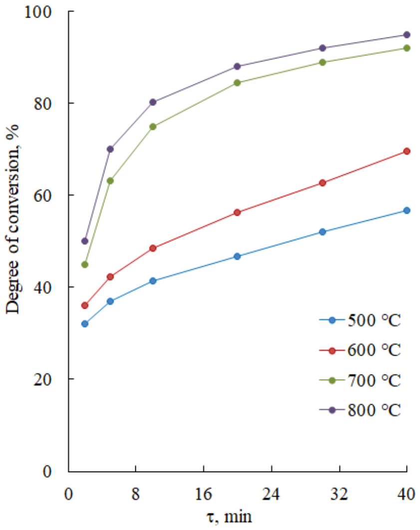

Figure 7. Dependence of the degree of conversion on time for system II ($PbO \cdot PbCl_2$–Na_2CO_3–C) at various temperatures

To evaluate changes in the reaction mechanism, the apparent activation energy was analyzed at degrees of conversion of α = 45% and α = 55%. The obtained values are presented in Table 2 and illustrated in Figure 8.

Table 2. Apparent activation energy values for the interaction of $PbO \cdot PbCl_2$ with Na_2CO_3 and C at different degrees of conversion

Degree of conversion α, %	Temperature range, °C	E_a, kJ/mol
45	500-600	43.5
45	600-700	43.5
45	700-800	44.0
55	500-600	64.5
55	600-700	66.5
55	700-800	74.0

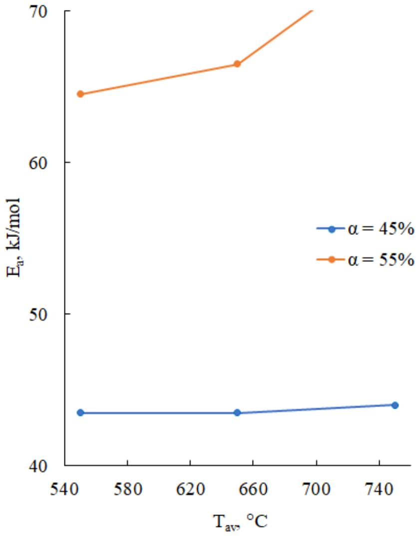

Figure 8. Dependence of the apparent activation energy (E_a) on the average temperature of the temperature interval (T_{av}) for system II ($PbO \cdot PbCl_2$–Na_2CO_3–C) at degrees of conversion α = 45% and α = 55%

It was established that at α = 45%, the apparent activation energy remains nearly constant, amounting to 43.5-44.0 kJ/mol throughout the entire investigated temperature range, which is characteristic of a predominantly chemically controlled reaction regime. As the degree of conversion increases to α = 55%, the apparent activation energy rises from 64.5 to 74.0 kJ/mol with increasing temperature. This indicates a complication of the reaction mechanism and an increasing influence of structural and diffusion limitations typical of multistage topochemical reactions [16].

The increase in activation energy values with increasing degree of conversion confirms the multistage nature of the reductive interaction of lead oxychloride with sodium carbonate and carbon. The initial stage of the reaction proceeds predominantly under chemical control, whereas at later stages mass transfer processes and the formation of a dense product layer begin to play a significant role, hindering the further progression of the reaction.

3.3. Comparative analysis of kinetic behavior in systems I and II

A comparative analysis of the kinetic behavior of systems I ($PbCl_2$-Na_2CO_3-C) and II ($PbO \cdot PbCl_2$-Na_2CO_3-C) reveals significant differences in reaction rate, kinetic regime, and limiting stages of the reduction process, despite the use of the same sodium-based fluxing additive. These differences are primarily associated with the chemical nature of the lead-containing compounds and the presence of oxygen in the structure of lead oxychloride.

In system I, the reduction of lead chloride proceeds relatively slowly and allows a clear identification of the initial stage governed by chemical kinetics. The experimental data demonstrate that at low degrees of conversion the process is satisfactorily described by a first-order kinetic model, enabling the determination of the apparent rate constant and subsequent Arrhenius analysis. The relatively low apparent activation energy at the initial stage indicates that the reaction rate is mainly controlled by chemical interaction between $PbCl_2$, Na_2CO_3, and carbon. As the reaction proceeds, the accumulation of reaction products and changes in the solid-phase structure lead to a transition toward a mixed kinetic regime, accompanied by an increase in the apparent activation energy, which is consistent with general kinetic concepts of extractive metallurgy and solid-state reaction theory [15-18].

In contrast, system II is characterized by a substantially higher reaction rate and rapid attainment of high degrees of conversion, even at moderate temperatures. The presence of oxygen in the $PbO \cdot PbCl_2$ structure facilitates the decomposition of the compound and promotes faster interaction with sodium carbonate and the reducing agent. As a result, the initial chemically controlled stage is significantly shortened, and the reliable determination of the rate constant based on a first-order kinetic model becomes impractical. For this reason, the kinetic analysis of system II was performed using the apparent activation energy determined at fixed degrees of conversion.

The comparison of apparent activation energies further highlights the fundamental differences between the two systems. For system I, the activation energy at the initial stage is relatively low and increases progressively with conversion, reflecting the gradual transition from chemical control to a regime influenced by mass transfer and structural factors. In system II, the activation energy at moderate conversion levels ($\alpha \approx$ 45%) remains nearly constant over the investigated temperature range, indicating a predominantly chemically controlled process. However, at higher degrees of conversion ($\alpha \approx$ 55%), a pronounced increase in the apparent activation energy is observed, which points to the growing influence of diffusion limitations and the formation of a dense product layer.

The thermodynamic analysis confirmed that both reactions are feasible over the studied temperature range and exhibit a significant driving force. Therefore, the observed differences in reaction behavior are primarily associated with kinetic factors rather than thermodynamic limitations.

Overall, the comparative analysis demonstrates that the introduction of an oxygen-containing lead compound substantially alters the kinetic characteristics of the reduction process in sodium fluxing systems. While both systems ultimately exhibit a multistage reaction mechanism, the relative contribution of chemical and diffusion-controlled stages differs markedly. These findings are of practical importance for optimizing flux composition and operating conditions in the processing of lead-containing dusts, particularly with regard to achieving high metal recovery while maintaining process efficiency.

4. Conclusions

Thermodynamic analysis demonstrated that both reduction reactions are feasible in the temperature range 500–1000°C and are characterized by a substantial driving force.

It was established that the reductive interaction of lead chloride and lead oxychloride with sodium carbonate and carbon proceeds via a multistage mechanism accompanied by a change in the kinetic regime as the reaction progresses.

For system I ($PbCl_2$-Na_2CO_3-C), the initial stage of the process is satisfactorily described by first-order kinetics, indicating the predominance of chemical control and allowing reliable determination of the rate constant and apparent activation energy.

System II ($PbO{\cdot}PbCl_2$-Na_2CO_3-C) exhibits significantly higher reactivity and rapid attainment of high degrees of conversion, which substantially shortens the chemically controlled stage and limits the applicability of first-order kinetic analysis.

Analysis of the apparent activation energy values shows that, for system I, the increase in activation energy with increasing conversion reflects a gradual transition from chemical control to a regime influenced by mass transfer and structural changes of the solid phase, in agreement with kinetic models reported for carbothermic reduction of metallurgical dusts. In contrast, for system II, a predominantly chemically controlled regime is preserved at moderate conversion levels.

The obtained results demonstrate that the presence of oxygen in the structure of lead oxychloride has a pronounced effect on the reduction kinetics in sodium-based fluxing systems and should be considered when developing and optimizing technological schemes for the processing of lead-containing dusts.

Author contributions

Conceptualization: GZM, BSB; Data curation: SKJ, MDT; Formal analysis: YBT, MDT; Funding acquisition: GZM; Investigation: AAD, SKJ; Methodology: YBT, AAD; Project administration: GZM; Resources: AAI, GMK; Software: MDT, SKJ; Supervision: BSB, GZM; Validation: GMK, AAI, YBT; Visualization: MDT, SKJ, GMK; Writing – original draft: GZM, BSB, AAD; Writing – review & editing: YBT, GMK, SKJ. All authors have read and agreed to the published version of the manuscript.

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Conflicts of interests

The authors declare no conflict of interest.

Data availability statement

The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

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Қорғасын хлориді мен оксихлоридінің натрий карбонатымен тотықсыздану кинетикасы

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Аңдатпа. Осы мақалада көміртектің қатысуымен қорғасын хлориді ($PbCl_2$) мен қорғасын оксихлоридінің ($PbO \cdot PbCl_2$) натрий карбонатымен тотықсыздана әрекеттесуінің кинетикалық заңдылықтары зерттелді. Зерттеудің өзектілігі екінші реттік өндірістен алынатын қорғасынқұрамды шаңдарды өңдеудің тиімді технологияларын әзірлеу және натрийқұрамды флюстердің құрамын оңтайландыру қажеттілігімен негізделеді. Кинетикалық зерттеулер $PbCl_2$–Na_2CO_3–C жүйесі үшін 800-1100°C және $PbO \cdot PbCl_2$–Na_2CO_3–C жүйесі үшін 500-800ºC температура аралығында изотермиялық жағдайларда термогравиметриялық әдіспен жүргізілді. Тотықсыздана әрекеттесу процесі түрлену дәрежесінің артуына қарай шектейтін сатының өзгеруімен сипатталатын көпсатылы механизм бойынша жүретіні анықталды. $PbCl_2$–Na_2CO_3–C жүйесі үшін процестің бастапқы бөлігі бірінші реттік реакция теңдеуімен қанағаттанарлық сипатталады, бұл химиялық бақылаудың басым екенін көрсетеді; осы сатыдағы көрінетін энергия активациясы 33.5 кДж/мольді құрайды. Түрлену дәрежесінің артуымен энергия активациясының 62-80 кДж/мольге дейін өсуі байқалады, бұл процестің диффузиялық және құрылымдық шектеулермен күрделенген режимге ауысуын көрсетеді. $PbO \cdot PbCl_2$–Na_2CO_3–C жүйесі жоғары реакциялық қабілетімен және түрленудің едәуір дәрежелеріне жылдам жетуімен сипатталады. Орташа түрлену дәрежелерінде процесс негізінен химиялық бақылау жағдайында жүреді ($E_a \approx$ 43-44 кДж/моль), алайда $\alpha \geq 55\%$ кезінде энергия активациясының 74 кДж/мольге дейін өсуі байқалады, бұл массаалмасу әсерінің күшейетінін көрсетеді. Қорғасын оксихлоридінің құрылымындағы оттегінің болуы натрийқұрамды флюстік жүйелердегі тотықсыздандыру процесінің кинетикалық сипаттамаларына елеулі әсер ететіні көрсетілді. Алынған нәтижелер қорғасынқұрамды шаңдарды өңдеудің технологиялық схемаларын оңтайландыруда, металды шығару дәрежесін арттыру және процестің энергия тиімділігін жоғарылату мақсатында қолданылуы мүмкін.

Негізгі сөздер: *қорғасын құрамды шаң, қорғасын хлориді, натрий карбонаты, тотықсыздандырып электрбалқыту, қатты фазалы реакциялар кинетикасы, энергия активациясы, термогравиметриялық талдау*.

Кинетика восстановления хлорида и оксихлорида свинца карбонатом натрия

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Аннотация. В работе исследованы кинетические закономерности восстановительного взаимодействия хлорида свинца ($PbCl_2$) и оксихлорида свинца ($PbO \cdot PbCl_2$) с карбонатом натрия в присутствии углерода. Актуальность исследования обусловлена необходимостью разработки эффективных технологий переработки свинецсодержащих пылей вторичного производства и оптимизации состава натрийсодержащих флюсов. Кинетические исследования выполнены термогравиметрическим методом в изотермических условиях в диапазоне температур 800-1100°C для системы $PbCl_2$–

Na_2CO_3–C и 500–800°C для системы $PbO{\cdot}PbCl_2$–Na_2CO_3–C. Установлено, что восстановительное взаимодействие протекает по многостадийному механизму с изменением лимитирующей стадии по мере увеличения степени превращения. Для системы $PbCl_2$–Na_2CO_3–C начальный участок процесса удовлетворительно описывается уравнением реакции первого порядка, что свидетельствует о преобладании химического контроля; кажущаяся энергия активации на этой стадии составляет 33.5 кДж/моль. С увеличением степени превращения наблюдается рост энергии активации до 62-80 кДж/моль, что указывает на переход к режиму, осложнённому диффузионными и структурными ограничениями. Система $PbO{\cdot}PbCl_2$–Na_2CO_3–C характеризуется более высокой реакционной способностью и быстрым достижением значительных степеней превращения. При умеренных степенях превращения процесс протекает преимущественно в условиях химического контроля ($E_a \approx$ 43-44 кДж/моль), однако при $\alpha \geq 55\%$ наблюдается увеличение энергии активации до 74 кДж/моль, свидетельствующее о возрастании влияния массопереноса. Показано, что присутствие кислорода в структуре оксихлорида свинца существенно влияет на кинетические характеристики восстановительного процесса в натрийсодержащих флюсовых системах. Полученные результаты могут быть использованы при оптимизации технологических схем переработки свинецсодержащих пылей с целью повышения извлечения металла и энергоэффективности процесса.

Ключевые слова: *свинецсодержащая пыль, хлорид свинца, оксихлорид свинца, карбонат натрия, восстановительная электроплавка, кинетика твёрдофазных реакций, энергия активации, термогравиметрический анализ.*

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