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Sakarya University Journal of Science
SAUJS

e-ISSN 2147-835X Period Bimonthly Founded 1997 Publisher Sakarya University
http://www.saujs.sakarya.edu.tr/

Title: Electrochemical Reduction of Lead Sulfide in NaCl-KCl and NaCl-KCl-%2Na2S

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Recieved: 2021-07-16 00:00:00

Accepted: 2022-02-08 00:00:00

Article Type: Research Article

Volume: 26
Issue: 2
Month: April
Year: 2022
Pages: 232-240

How to cite
Levent KARTAL; (2022), Electrochemical Reduction of Lead Sulfide in NaCl-KCl and NaCl-KCl-%2Na2S. Sakarya University Journal of Science, 26(2), 232-240, DOI: 10.16984/saufenbilder.972380
Access link
https://dergipark.org.tr/tr/journal/1115/issue/69580/972380

Sakarya University Journal of Science 26(2), 232-240, 2022

Electrochemical Reduction of Lead Sulfide in NaCl-KCl and NaCl-KCl-%2Na_2S

Levent KARTAL*[1]

Abstract

In this study, the lead production from lead sulfide (PbS) by molten salt electrolysis was investigated under potentiostatic and galvanostatic conditions using NaCl-KCl and NaCl-KCl-2%Na_2S electrolytes. Stable cell voltage and current were aimed with the addition of Na_2S to the NaCl-KCl electrolyte. Reduction experiments were carried out at constant 700 °C temperature and for 15 min. duration. The current density was set to 250 mA/cm^2 for the galvanostatic reduction experiments. It was observed that there was an increase in cell voltage in both electrolytes due to the decrease in the amount of PbS in galvanostatic experiments. In these experiments, it was determined that the reduction occurred at higher cell voltages in the NaCl-KCl electrolyte compared to the NaCl-KCl-2%Na_2S electrolyte. Although the cell voltage was aimed to remain constant with the Na_2S addition, the cell voltage decreased slightly compared to the NaCl-KCl electrolyte, but increased with the experiment duration as in the NaCl-KCl electrolyte. Potentiostatic reduction experiments carried out at a constant cell voltage of 3.0 V under the electrolyte decomposition voltage. The morphology of the cathode products was examined by SEM-EDS analysis and, the phases were examined by X-ray diffractometry. Higher current values were obtained in the NaCl-KCl-%2Na_2S electrolyte compared to the NaCl-KCl electrolyte. Current variation with electrolysis duration showed similar trends in both electrolytes. According to structural characterization, it was determined that the metallic lead mass did not contain any impurities.

Keywords: Molten salt electrolysis, electrochemical-reduction, lead extraction, lead sulfide

1. INTRODUCTION

Lead has been widely used by mankind for many years due to its features such as ease of production, low melting temperature, good formability, and high corrosion resistance. With the determination of its toxic effects thanks to the developing technology, the direct or indirect use of lead in many products have been banned or limited. However, it is still widely used in the battery manufacturing industry, insulation of underground communication cables, protection from X-rays due to its radiation-blocking properties, and ammunition production [1, 2].

The most common lead sources are galena (PbS), serousite ($PbCO_3$) ores, and waste batteries. Primary lead production is made from galena (PbS) concentrates by pyrometallurgical methods and is refined by pyrometallurgical/electrometallurgical processes and presented to the industry [3, 4]. Pyrometallurgical production is carried out in

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systems where direct melting processes (QSL, Ausmelt, Outokumpu, etc.), oxidation and reduction stages take place in the same furnace. These processes were developed to reduce the use of coke by utilizing the oxidation heat of the sulfur in the concentrate and to eliminate the costs of expensive gas cleaning, dust holding and ventilation equipment [5, 6].

In addition to oxygen and sulfur, lead concentrates generally contain iron, copper, nickel, cobalt, zinc, arsenic, antimony, tin, bismuth, selenium, tellurium, silver, gold, and these metals are collected in lead after reduction. Purification of lead from these metals is done by multi-stage pyrometallurgical, and electrometallurgical refining steps. In pyrometallurgical lead refining, copper is removed by adding S to lead, forming copper sulfide (Cu_xS_y) and withdrawing it from the surface, removal of As, Sb, and Sn by adding sodium nitrate ($NaNO_3$) to molten lead, and separating it to form sodium arsenate, antimonite, and sodium stannate compounds. Removal of Zn is accomplished by oxidation, chlorination or evaporation under vacuum. Electrometallurgical lead refining is achieved by dissolving lead anodes in fluoboric, fluosilicic and sulfamic acid-based electrolytes and depositing it at the cathode at an appropriate current density. In refining electrolysis, fluosilicic acid is generally preferred as the electrolyte due to its low price [6, 7].

Existing metallurgical processes used in metal production from sulfide ores are processes with high initial investment and operating cost. In addition, SO_2, CO, and CO_2 released in the environment during production with the traditional methods cause serious environmental problems [8]. Increasing environmental restrictions and expensive SO_2 capture systems urge researchers to develop both environmental and economic methods for sustainable metal production. The development of systems that provide stable sulfide recovery in elemental form along with metal production is of environmental and economic importance. In the production of metal from metal sulfide compounds, the most emphasized methods for the production of metal and elemental sulfur directly from metal sulfide are hydrometallurgical and electrochemical production methods. Since too much acid is consumed in the hydrometallurgical method, S forms H_2S, and an oxidant is needed to obtain elemental sulfur, it has not yet reached the stage to replace pyrometallurgical production. Although direct metal production from oxide or sulfide compounds by electrometallurgical methods has been known for many years, it gained momentum in the late 90’s with the study of a group of researchers from Cambridge University on the electrochemical reduction of titanium dioxide [9]. In the process, the direct reduction of oxidized/sulfurous compounds by molten salt electrolysis, the compounds are cathodically polarized in suitable electrolytes and metal formation is provided, while the ionized O^{2-}/S^{2-} reaches the anode (usually graphite) where it is oxidized to form S_2, CO, CO_2 gases [10]. In molten salt electrolysis; electrolyte, anode material, and applied temperature are the most important components. The electrolyte is of great importance as it affects the working temperature with ion transport, and the melting temperature. In literature, NaCl-KCl is used as an electrolyte due to high S^{2-} diffusion in the reduction of sulfide compounds, while the use of $CaCl_2$ is prominent in the reduction of oxide compounds due to high O^{2-} diffusion [11, 12]. As the anode, carbon-based electrodes that are easy to mass-produce and supply are preferred. While these electrodes cause greenhouse gas (CO, CO_2) emission in the reduction of oxide compounds, carbon-based anodes act inert in the reduction of sulfide ores. Thus, with the formation of metal at the cathode and sulfur at the anode, metal production is possible with the use of a cheap anode without greenhouse gas emissions [13].

In studies on lead production by molten salt electrolysis, there are studies with chloride (LiCl-KCl), fluoridated (HBF_4), and hydroxide (NaOH) electrolytes. In chloride electrolytes, lead production was carried out at 450 °C using the LiCl-KCl-$PbCl_2$ electrolyte with graphite electrodes [14]. Doe Run and Engitec companies have developed the Engitec Flubor molten salt electrolysis process using HBF_4 electrolyte for lead generation. In the process, lead is based on ferric leaching and electrochemical reduction of galena concentrate in a fluoroboric acid-based solution. Electro-winning is carried out in split cells using stainless steel cathodes and graphite anodes [15]. On the other hand, Buzatu et al., stated that they performed their studies in the production of Pb from wastes containing $PbSO_4$, PbO, and Pb in a NaOH based electrolyte, using stainless steel anodes and cathodes, at a current density of 600 A/m^2, at an electrolyte temperature of 35 °C [16]. Lead in powder form production at low temperature without leaching was studied by Qu et al. They stated that it is possible to produce powdered lead in 50% NaOH solutions at 120 °C, which they pelletized PbS powders [17].

In this study, lead production was carried out in one step under galvanostatic, and potentiostatic conditions at a constant temperature of 700 °C in NaCl-KCl and NaCl-KCl-2%Na_2S electrolytes. Castable lead production, which does not require extra leaching of lead sulfide and production of intermediate chloride compounds from lead sulfide, was investigated. In addition, the effects of Na_2S added to the electrolyte on cell voltage, anode, and cathode potentials under galvanostatic conditions and on current under potentiostatic conditions were examined.

2. EXPERIMENTAL

The materials used in this study are given in Table 1 along with their technical specifications.

Table 1 Materials used in electrochemical reduction experiments

Materials	Form	Specification
PbS	Powder	technical grade purity
NaCl	Powder	technical grade purity
KCl	Powder	technical grade purity
Na_2S	Powder	technical grade purity
Cathode	Graphite crucible	Ø 45 mm, h 100 mm
Anode	Graphite rod	ϕ 16 mm

NaCl-KCl constituting the electrolyte was prepared by mixing in 1:1 mole ratio considering the phase diagram. Salt mixtures, graphite crucible (65mmOD, 45mmID, 100mm height), and other graphite materials were kept in an oven at 110 °C for moisture removal. Argon gas (50 ml/min) was used as a protective atmosphere in all experiments. Pre-electrolysis was performed for 1 hour by applying a 2.7 V constant cell potential to remove electrolyte-sourced impurities. Graphite rods with a diameter of 6 mm were used as reference electrodes in the experiments. In the galvanostatic and potentiostatic reduction experiments, the heating was carried out using a medium frequency induction furnace (50 kHz, 30 kW, 40 A). In these experiments, 16 mm diameter graphite rods were used as the anode. The anode height was determined as 20 mm from the bottom of the crucible, taking into account the densities of the materials fed to the cell (electrolyte (2.05 g/cm^3), lead sulfide (7.6 g/cm^3), and lead (11.34 g/cm^3). The anode was positioned on the lead sulfide in the electrolyte to prevent short circuit formation. Fixed 3 g of PbS was used in all reduction experiments.

Characterization studies were carried out using optical emission spectrometry (OES, GNR-S7 MLP), X-ray diffractometer (XRD, Siemens D5000), scanning electron microscope with energy dispersive X-ray spectrometry (Jeol-JSM 5410).

3. RESULTS AND DISCUSSION

3.1. Electrochemical reduction of PbS

The breakdown voltages of lead sulfide (PbS) and the salts forming the electrolyte were calculated using theoretical data. Equation 1 was used to calculate the breakdown voltages (ΔE) and the values are given in Table 2. In Equation 1, n represents the number of electrons and F represents Faraday's constant. Since the decomposition potential of lead sulfide is much lower than the decomposition voltage of NaCl, KCl, and Na_2S used in electrolyte preparation, lead sulfide reduction takes place without chlorine (Cl_2) emission.

$$\Delta E = -\Delta G / (nF) \ [18] \qquad (1)$$

Table 2 The standard Gibbs free energy changes along with decomposition voltages of PbS and NaCl, KCl at 700 °C

Reaction	Gibbs Free Energy Changes $\Delta G°$/kJ mol^{-1}	Theoretical Decomposition Voltage/V
$2NaCl = 2Na + Cl_2(g)$	642.405	-3.33
$2KCl = 2K + Cl_2(g)$	688.058	-3.56
$2Na_2S=4Na+S_2(g)$	616.952	-1.6
$2PbS = 2Pb + S_2(g)$	154.416	-0.40

The variation of the standard Gibbs free energy of formation of the materials used in experimental studies with temperature is shown in figure 1. Based on these data, the variation of the decomposition potentials with temperature is given in figure 2.

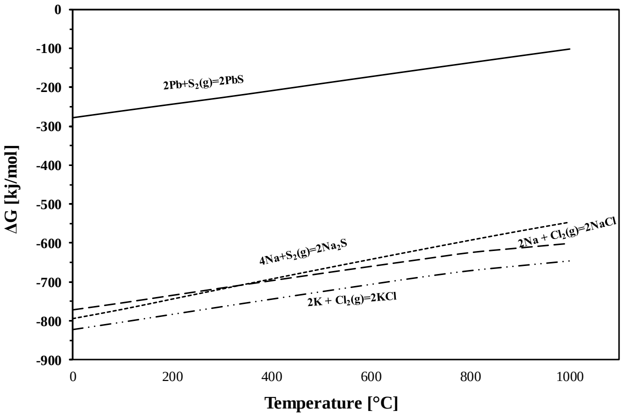

Figure 1 Change in the standard Gibbs free energy of formation of PbS, NaCl, KCl, and Na_2S as a function of temperature

It is understood that the decomposition potentials of PbS, and Na_2S, which are expected to be reduced under galvanostatic/potentiostatic experimental conditions, are much lower than the NaCl, and KCl constituting the electrolyte so that lead production can occur without harmful gas emissions (fig 2).

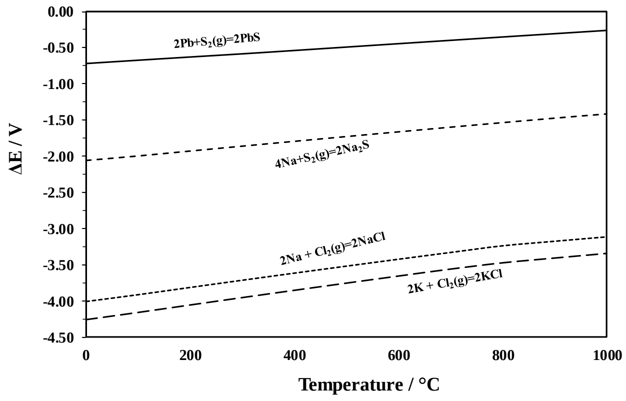

Figure 2 The decomposition potentials (ΔE°) for PbS, NaCl, KCl, and Na_2S as a function of temperature

The expected anode and cathode reactions as a result of thermodynamic investigations and calculations are given below.

$$\text{Cathode} \quad : PbS+2e^- = Pb+S^{2-} \qquad (2)$$

$$\text{Anode} \quad : S^{2-}-2e^- = 1/2S_2(g) \qquad (3)$$

$$\text{Net Reaction} \quad : PbS = Pb+1/2S_2(g) \qquad (4)$$

The lead accumulated in the cathode will collect at the bottom of the graphite crucible as a liquid and ready for casting due to the experimental operating temperature being higher than the melting temperature of lead. It is estimated that the sulfur formed at the anode leaves the system as a gas due to the operating temperature being higher than the evaporation temperature of the sulfur and condenses in cold regions.

3.2. Galvanostatic Electrolysis of PbS

The galvanostatic reduction experiments of PbS were carried out at the constant temperature of 700 °C using two different electrolytes, NaCl-KCl and NaCl-KCl-2% Na_2S. By adding Na_2S to the NaCl-KCl electrolyte, a saturation of the electrolyte with sulfur and stable cell potential was aimed. In both electrolytes, it was determined that the cell potential increased over time due to the decrease in the amount of PbS that decreased with the electrolysis time. It was observed that the cell voltage increase was sharper in the NaCl-KCl electrolyte compared to the NaCl-KCl-2% Na_2S electrolyte and almost reached the electrolyte breakdown potential in a short experiment time of 15 minutes. Although the cell voltage in the electrolyte using NaCl-KCl-%2Na_2S was lower than the NaCl-KCl electrolyte, it started to increase at the end of the experiment due to the decrease in the amount of PbS. Yin et al. studied Sb2S3 reduction in NaCl-KCl-Na2S electrolyte and observed that the cell voltage increased from about 0.5 V to 2.25 V for 45 minutes [21].

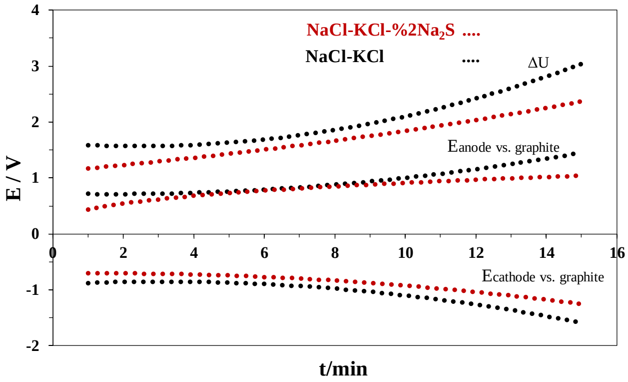

Figure 3 Variations of cell voltage at 250 mA/cm^2 in NaCl−KCl−PbS, and NaCl−KCl−%2 Na_2S-PbS melt at 700 °C

The chemical content of the cathode products produced in both electrolytes was investigated by an OES. It was observed that the lead obtained as a result of both studies did not contain any metallic impurities and high amount of sulfur residues (Table 3).

Table 3 OES results for cathode products

Electrolytes	The concentration of element *i*, *Ci* (wt.%)	
	Pb	S
NaCl−KCl	99.04	0.6
NaCl−KCl−%2 Na_2S	99.07	0.5

The products produced using NaCl−KCl, and NaCl−KCl−%2 Na_2S electrolytes were examined by SEM-EDS analysis and no impurities were detected in the metallic mass.

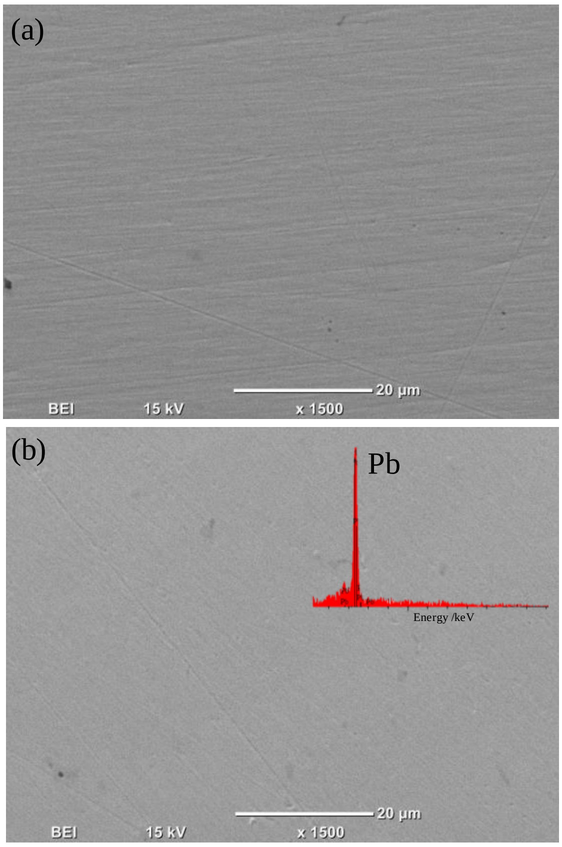

Figure 4 SEM images of the cathode products produced in (a) NaCl-KCl and (b)NaCl-KCl-Na_2S for 15 min. at 250 mA/cm^2 at 700 °C

In Figure 5, X-ray analysis results of lead produced in both electrolytes are given. It has been confirmed that the results of X-ray analysis support the results of OES and SEM-EDS analysis and that there are no oxidized/sulfurous phases in the structure. In accordance with the work of Yin et al., high purity metallic lead was obtained in the NaCl-KCl-Na_2S based electrolyte [21].

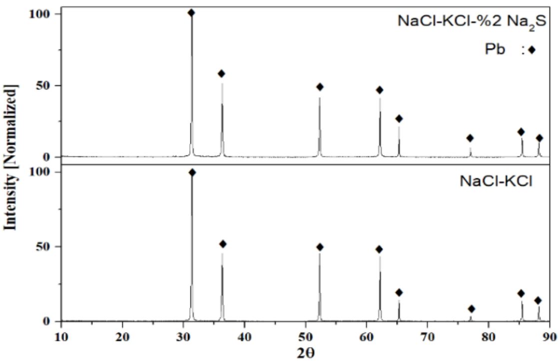

Figure 5 XRD patterns of Pb synthesized at 250 mA/cm^2 in (a) NaCl-KCl, and (b)NaCl-KCl-Na_2S melt at 700 °C

3.3. Potentiostastic Electrolysis of PbS

Lead production from PbS was also investigated under potentiostatic conditions using the same electrolytes, due to the rapid increase in the cell potential in the experiments performed under galvanostatic conditions and approaching decomposition potential of the chloride salts forming the electrolyte. Taking into account the theoretical calculations (see Table 2 and figures 1, 2), the experiments were carried out at a constant cell voltage of 3.0 V, under the breakdown voltage of the salts forming electrolyte and above the breakdown voltage of PbS. Figure 6 shows the current changes depending on the electrolysis time at a fixed cell potential of 3.0 V. The current changes obtained as a result of the experiments carried out at constant potential in two different electrolytes show a similar trend and the currents became stable after approximately 10 minutes. Qu et al. has investigated the electrochemical lead production from PbS in 50% NaOH water solution under constant voltage at 120 °C. The current behavior, similar to the study of Qu et al., the current decreased rapidly over time and continued as a flat plateau [17]. Li et al. studied the electrochemical reduction of Sb_2S_3-PbS in NaCl-KCl at 730 °C at different cell voltages [22]. Although the current values in Li and co-workers study were very different, the trend was similar to the results of Qu. The current was higher in the electrolyte containing Na_2S compared to the electrolyte without Na_2S.

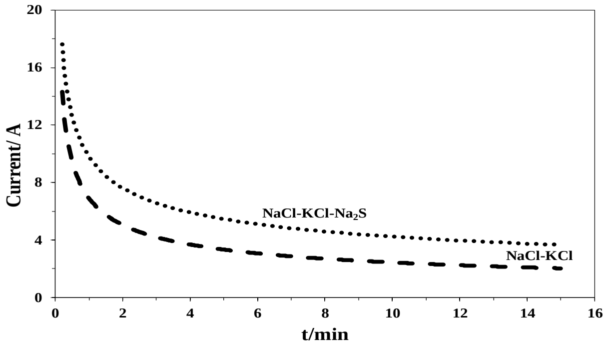

Figure 6 Current versus time at different voltages in NaCl−KCl−PbS and NaCl−KCl−%2Na_2S-PbS melt at 3.0 V, and 700 °C

The chemical content of the cathode products produced under potentiostatic conditions was examined by OES and was found that it did not contain high sulfur residues with a lead content of over 99 % (Table 4).

Table 4 Chemical analysis of cathode products

Electrolytes	The concentration of element *i*, *Ci* (wt.%)	
	Pb	S
NaCl−KCl	98.7	<0.1
NaCl−KCl−%2 Na_2S	99.2	<0.1

The obtained metallic lead was examined by SEM-EDS, and it was seen that the black spots on the surface were not sulfide residues and impurities but voids. XRD and SEM-EDS analyses also support the OES results (fig. 8, 9). Li et al. studied the electrochemical reduction of Sb_2S_3-PbS in NaCl-KCl and it is understood from XRD studies that in high purity Pb is produced at 2.2 V. The sulfide structures in metallic lead were removed with increasing time [22]. Qu et al. investigated the electrochemical lead production from PbS at different cell voltages[17]. Similar to the work of Li et al., Qu et al. also obtained high purity lead at 2.0 V and above. The highest possible voltage under the electrolyte breakdown voltage shortens the reduction time. Therefore, instead of trying different voltages, 3.0 V was applied in our study. This result is also supported by the studies of Qu and Li.

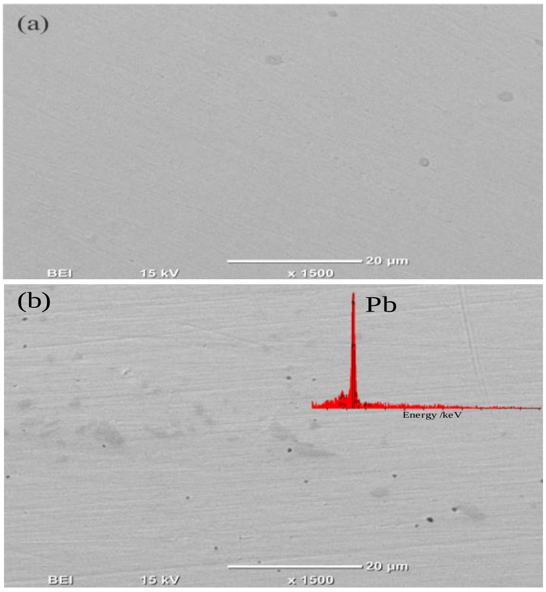

Figure 7 SEM images of the cathode products produced in (a) NaCl-KCl, and (b)NaCl-KCl-%2 Na_2S for 15 min. at 3.0 V at 700 °C

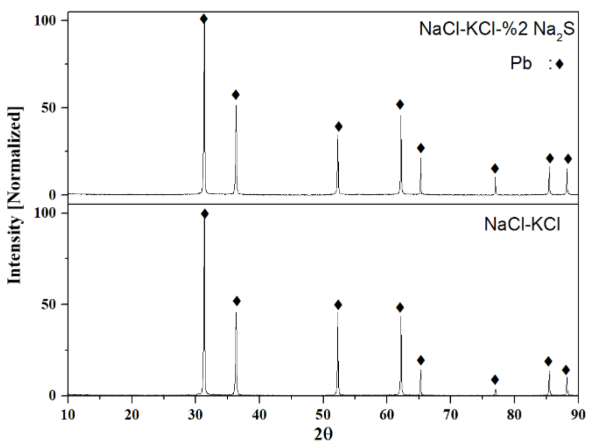

Figure 8 XRD patterns of Pb synthesized at 3.0 V in NaCl−KCl−PbS and NaCl−KCl−%2 Na_2S-PbS melt at 700 °C

Graphite is an ideal inexhaustible anode material for sulfur production as a result of the anodic oxidation of S^{2-}. This has been proven by numerous studies on the electrolysis of solid metal sulfides and dissolved metal sulfides [11, 19, 20]. The oxidized sulfur on the anode leaves the system as a gas at an operating temperature of 700 °C. While a significant part of the sulfur formed at the anode is kept in gas scrubbing bottles and it has been determined that some of it condense in cold regions.

4. CONCLUSION

In this study, lead production was investigated from lead sulfide (PbS) without SO_2 emission in NaCl-KCl, and NaCl-KCl-%2 Na_2S electrolyte at 700 °C. The key conclusions can be summarized as follows;

- In the experiments performed with NaCl-KCl electrolyte at a current density of 250 mA/cm^2 under galvanostatic conditions, it was determined that the cell voltage increased due to the decrease in the amount of PbS and reached the electrolyte breakdown voltage.
- In NaCl-KCl-%2Na_2S electrolyte, the Na_2S addition could not provide stable cell voltage, the cell potential increased over time, but the increase was slower than the NaCl-KCl electrolyte.
- In the trials carried out by applying constant 3.0 V in NaCl-KCl and NaCl-KCl-%2Na_2S electrolytes, it was observed that the current change over time was similar, and the current values were higher in the electrolyte containing Na_2S.
- Stable cell voltage and current values could not be obtained under galvanostatic and potentiostatic conditions with the addition of Na_2S to the electrolyte.
- The sulfur formed at the anode during electrolysis was collected in gas scrubbing bottles and in the cold areas of the cell.

Acknowledgments

The authors would like to acknowledge the reviewers and editors of Sakarya University Journal of Science.

Funding

The author (s) has no received any financial support for the research, authorship or publication of this study.

The Declaration of Conflict of Interest/ Common Interest

No conflict of interest or common interest has been declared by the authors.

The Declaration of Ethics Committee Approval

This study does not require ethics committee permission or any special permission.

The Declaration of Research and Publication Ethics

The authors of the paper declare that they comply with the scientific, ethical and quotation rules of SAUJS in all processes of the paper and that they do not make any falsification on the data collected. In addition, they declare that Sakarya University Journal of Science and its editorial board have no responsibility for any ethical violations that may be encountered, and that this study has not been evaluated in any academic publication environment other than Sakarya University Journal of Science.

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