

## Research Article

# A Method for the Simultaneous Cleansing of H<sub>2</sub>S and SO<sub>2</sub>

Dzhamal R. Uzun,<sup>1</sup> Elena Razkazova-Velkova,<sup>2</sup> Venko Beschkov,<sup>2</sup> and Konstantin Petrov<sup>1</sup>

<sup>1</sup>*Institute of Electrochemistry and Energy Systems “Acad. Evgeni Budevski”, Bulgarian Academy of Sciences, Acad. G. Bonchev Street, Building 10, 1113 Sofia, Bulgaria*

<sup>2</sup>*Institute of Chemical Engineering, Bulgarian Academy of Sciences, Acad. G. Bonchev Street, Building 103, 1113 Sofia, Bulgaria*

Correspondence should be addressed to Dzhamal R. Uzun; [dzhamal.uzun@gmail.com](mailto:dzhamal.uzun@gmail.com)

Received 4 September 2015; Revised 19 November 2015; Accepted 13 December 2015

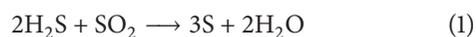
Academic Editor: Abel C. Chialvo

Copyright © 2016 Dzhamal R. Uzun et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

A method for the simultaneous electrochemical purification of hydrogen sulfide and sulfur dioxide from sea water or industrial wastes is proposed. Fundamentally the method is based on the electrochemical affinity of the pair H<sub>2</sub>S and SO<sub>2</sub>. The reactions (oxidation of H<sub>2</sub>S and reduction of SO<sub>2</sub>) proceed on a proper catalyst in a flow reactor, without an external power by electrochemical means. The partial curves of oxidation of H<sub>2</sub>S and reduction of SO<sub>2</sub> have been studied electrochemically on different catalysts. Following the additive principle the rate of the process has been found by intersection of the curves. The overall process rate has been studied in a flow type reactor. Similar values of the process rate have been found and these prove the electrochemical mechanism of the reactions. As a result the electrochemical method at adequate conditions is developed. The process is able to completely convert the initial reagents (concentrations C<sub>H<sub>2</sub>S, SO<sub>2</sub></sub> = 0), which is difficult given the chemical kinetics.

## 1. Introduction

A number of past methods for the removal of sulfur-containing compounds from gas and water streams have been investigated by different researchers. These methods generally separate the sulfur compounds from the gas stream by an absorption stage. The so-called Claus process is the most widely utilized [1] and follows the general reaction:



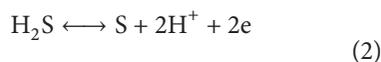
The Claus process proceeds at temperatures above 1300°C; however, by utilizing different catalysts, this can be reduced to 250–850°C [2–5]. The purification of SO<sub>2</sub> is currently a standalone process. There are different methods for extraction of sulfur dioxide, like neutralization methods, conversion of SO<sub>2</sub> into CaSO<sub>4</sub> [6], and liquefaction of SO<sub>2</sub> by adsorption in sulfate solution [7]. All these methods are based on the oxidation of SO<sub>2</sub>. Previous research by Petrov on the electrochemical oxidation reduction processes for SO<sub>2</sub> and H<sub>2</sub>S cleansing by their oxidation with oxygen contained in the air labeled ELCOX as innovative process for SO<sub>2</sub> cleansing in waste gases [8].

The present work is aimed at the opportunity of simultaneously cleansing the hydrogen sulfide and sulfur dioxide from industrial wastes and natural (Black Sea) H<sub>2</sub>S containing waters. It is well known that Black Sea deep waters contain an enormous amount of hydrogen sulfide (as hydrosulfides and sulfide ions) estimated as 4600 Tg, that is, 4.6 billion tons [9]. There are efforts to attack the problem by various methods: low temperature adsorption of hydrogen sulfide from sea waters followed by thermal decomposition [10, 11], electrolysis leading to hydrogen and sulfur production [12], or burning the hydrogen sulfide itself in electric power stations, as proposed by [13]. All these methods encounter the problem of low H<sub>2</sub>S concentrations (C<sub>H<sub>2</sub>S</sub> ~ 8–10 mg·L<sup>-1</sup>) which do not allow for the application of feasible technologies [14].

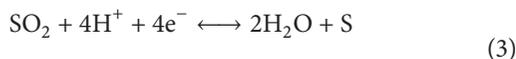
In this work we review the simultaneous reduction of SO<sub>2</sub> and oxidation of hydrogen sulfide. Various kinds of electrode materials were utilized in the past for the reduction of SO<sub>2</sub>, namely, mercury, gold, platinum, bismuth, copper, iron phthalocyanine on graphite, and uranium iron alloys. Previous studies do not fully explain the electrochemical process in the reduction of sulfur dioxide [15]. Various

electrochemical methods to convert hydrogen sulfide to its elements, at different concentrations, have been studied [16]. Different catalysts for the oxidation of hydrogen sulfide were used previously like ferric, cobalt, graphite, platinum, manganese cations, cobalt and carbon compounds, and perovskites or cobalt phthalocyanine [17–19]. The anodic process of sulfide oxidation can be carried out leading to different final products (elemental sulfur, polysulfides, or sulfates), depending on the reaction conditions: temperature, concentrations, catalysts, and so forth.

The aim of the proposed method is to simultaneously purify both reagents ( $\text{SO}_2$  and  $\text{H}_2\text{S}$ ) at room temperatures. Fundamentally the method is based on the electrochemical affinities of both compounds. The thermodynamic potentials of the pair  $\text{H}_2\text{S}$  and  $\text{SO}_2$  for oxidation and reduction are

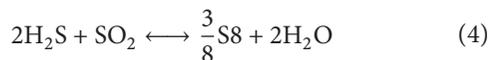


$$E_O = -0.14 \text{ V}$$



$$E_O = 0.17 \text{ V}$$

Overall reaction is



From the theory for concurrent redox (corrosion) reactions it follows that these electrochemical reactions can occur due to the potential difference of  $\Delta E \sim 0.30 \text{ V}$  as indicated for reactions (1) and (2) on dispersed microgalvanic elements [20, 21]. Based on this the reactions may proceed on the right catalyst in a flow reactor, without an external power source by electrochemical means. The reaction rate may be determined by studying the partial electrochemical reactions of  $\text{H}_2\text{S}$  oxidation and  $\text{SO}_2$  reduction, in accordance with the so-called additive principle [20–22].

An advantage of these electrochemical reactions is that they are able to completely convert the initial reagents (concentrations  $C_{\text{H}_2\text{S}, \text{SO}_2} = 0$ ) [23], which is difficult given the chemical kinetics [24]. An application has been filed for a patent entitled “the electrochemical method for separation of sulfur dioxide and hydrogen sulfide from fluids” [25].

The electrochemical realization of the process is possible due to the launching of novel microgalvanic cells. To illustrate the main idea a physical model of the process with a porous catalytic electrode is schematically shown in Figure 1. The model is similar to our previously published work [26]. The structure of the catalytic mass is composed of teflonized carbon black/charcoal with a deposited catalyst (1). Teflon is needed as a binding and hydrophobic agent because some of the reagents are in a gaseous form (e.g., 14% of  $\text{H}_2\text{S}$  in Black Sea waters exist as diluted gas). The catalytic particles are in direct electronic contact through a teflonized carbon network (2). Ion contact is achieved through the solution (electrolyte)—(3). These conditions are sufficient for the realization of a galvanic element between the pair  $\text{SO}_2$ - $\text{H}_2\text{S}$ . Depending on the pH of electrolyte the final products can be sulfur, polysulfides, or sulfates.

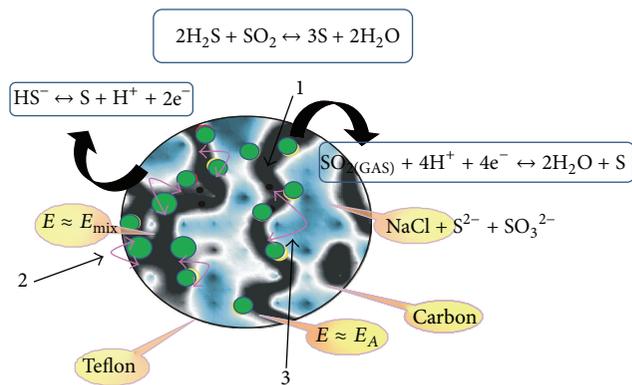


FIGURE 1: The physical model of a pore of a microgalvanic element with teflonized charcoal/carbon black and a catalytic mass for the reduction reaction of  $\text{SO}_2$  with the oxidation of  $\text{H}_2\text{S}$  in the electrolyte containing  $\text{H}_2\text{SO}_3 + \text{NaCl} + \text{NaOH} + \text{Na}_2\text{S} \cdot 9\text{H}_2\text{O}$ .

## 2. Materials and Methods

Sodium sulfide ( $\text{Na}_2\text{S} \cdot 9\text{H}_2\text{O}$ , Sigma-Aldrich 98.0%), Sodium sulfite ( $\text{Na}_2\text{SO}_3$ , Sigma-Aldrich), and Sodium Chloride ( $\text{NaCl}$ , Sigma-Aldrich) were for realizing of electrolyte. Deionized water was used. The Vulcan XC-72 carbon black (particle size  $\sim 50 \text{ nm}$ ) and acetylene blacks were from Cabot Corporation. The CoPc (cobalt phthalocyanine) was prepared by pyrolysis [18].

The total amount of sulfide and sulfite ions was determined iodometrically with starch as the indicator [27]. Sulfides were separately determined photometrically with N,N-dimethyl-n-phenylenediamine in the presence of Fe(III) and the formation of methylene blue [28–30]. This method allows for a doubled increase in the sensitivity of determination. The presence of thiosulfates, sulfites, and sulfates was also qualitatively checked. Thiosulfates form an unstable purple complex with Fe(III). The opalescence of the solution after the addition of barium cations shows the presence of sulfite and sulfate. The dissolution of the deposit in concentrated hydrochloric acid proves the presence of a sulfite, whereas the barium sulfate remains nondissolved [31].

We utilized a three-electrode cell to complete the electrochemical tests. For  $\text{SO}_2$  reduction we used sulphurous acid  $\text{H}_2\text{SO}_3$  ( $10 \text{ g} \cdot \text{L}^{-1}$ ). The  $\text{H}_2\text{S}$  concentration in the electrolyte has been varied in the range  $100\text{--}20\,000 \text{ mg} \cdot \text{L}^{-1}$  introduced as NaHS. This range has been selected having in mind that the electrochemical process is very slow (practically does not occur) at lower concentrations, especially at the naturally encountered concentration of about  $8 \text{ mg} \cdot \text{L}^{-1}$  [9], and higher concentrations were studied in previous investigations [14]. A supporting electrolyte of  $18 \text{ g} \cdot \text{L}^{-1}$  NaCl (similar to the concentration in Black Sea waters) is added to the electrolyte, as well as 1 M NaOH in some cases.

The cell volume is 150 mL. The electrolyte was stirred continuously with a magnetic stirrer. A Solartron 1286 Electrochemical Interface is used for the electrochemical measurements. A reference hydrogen electrode by “Gaskatel

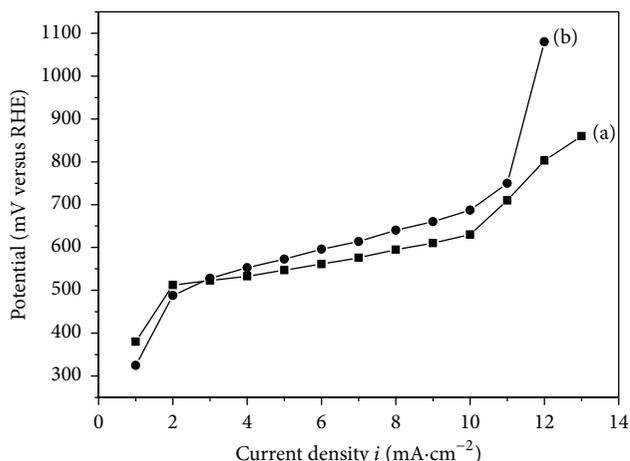


FIGURE 2: Working electrode potential versus current density for the optimized electrodes: ■ (a) DGCPV; ● (b) LSNV. Electrolyte— $1 \text{ g}\cdot\text{L}^{-1}$  sulfide ions;  $18 \text{ g}\cdot\text{L}^{-1}$  NaCl;  $1 \text{ M NaOH}$ ;  $t^\circ = 25^\circ\text{C}$ .

GmbH<sup>®</sup>, Germany, is applied. The counter electrode is Pt-foil. A minimum of four measurements were made for each result to achieve better reproducibility. Arithmetic averages are presented in the graphs.

An overall reaction rate for the simultaneous oxidation of hydrogen sulfide and reduction of sulfur dioxide without external power is studied in a flow type chemical reactor. The catalyst is placed in the reactor.

All the electrodes studied are of the immersed type and have a geometrical area of  $1 \text{ cm}^2$  or  $10 \text{ cm}^2$ . The electrodes are prepared from a mixture of the catalysts and teflonized carbon black (Vulcan XC-72 + 35% Teflon)—TV35—as a binder. The mixture is pressed onto both sides of a stainless steel current collector at  $t^\circ = 300^\circ\text{C}$  and  $P = 300 \text{ atm}$ .

### 3. Results and Discussion

**3.1. Electrode Properties and Optimization.** The immersed type electrodes for the oxidation of  $\text{H}_2\text{S}$  and reduction of  $\text{SO}_2$  were previously optimized with respect to the catalyst used, the electrode thickness, and the mass ratio between catalysts and binder by varying one parameter at a time and keeping the others constant [31, 32]. Different catalysts have been studied: (DG + 20% CoPc) + 35% teflonized Vulcan XC-72 (DGCPV); (DG + 20% CoPc) + 35% teflonized Acetylene Black (DCPAB);  $\text{La}_{1.3}\text{Sr}_{0.7}\text{NiO}_4$  + 35% teflonized Vulcan XC-72 (LSNV); Bulk Graphite; (DG + 20% CoPc) + 35% teflonized Vulcan XC-72 + 6% Pt., [31]. (DG + 20% CoPc) is active carbon type DG on which cobalt phthalocyanine is deposited [18].

The electrochemical characteristics of the best optimized electrodes for  $\text{H}_2\text{S}$  oxidation ((a) DG + 20% CoPc compare 35% teflonized Vulcan XC-72, DGCPV, ratio 30 : 70; the quantity is  $60 \text{ mg}/\text{cm}^2$ ; (b)  $\text{La}_{1.3}\text{Sr}_{0.7}\text{NiO}_4$  + 35% teflonized Vulcan XC-72, LSNV) are shown in Figure 2. It is seen that both electrodes provide fairly similar electrochemical behaviour.

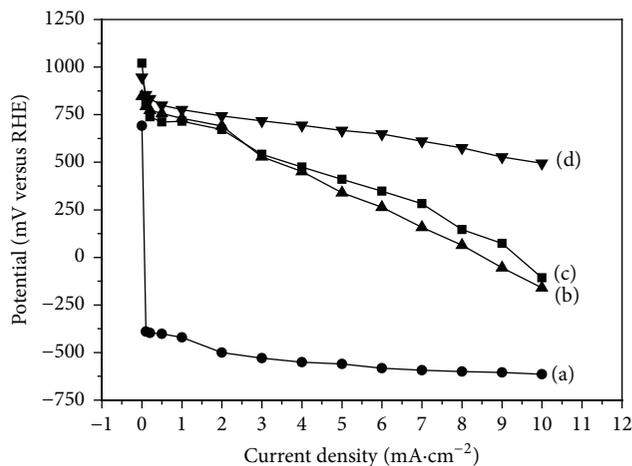


FIGURE 3: The polarization curves of sulfur dioxide reduction in an aqueous electrolyte:  $10 \text{ g}\cdot\text{L}^{-1}$   $\text{H}_2\text{SO}_3$ ;  $1 \text{ M NaOH}$ ;  $18 \text{ g}\cdot\text{L}^{-1}$  NaCl; (a) stainless steel; (b) TV35 and DG + 20% CoPc; (c) LSNV; (d) TV35 and DG + 20% CoPc + 6% Pt;  $t^\circ = 25^\circ\text{C}$ .

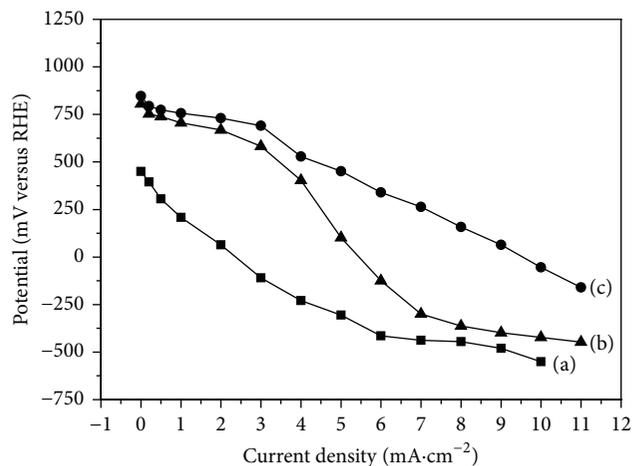


FIGURE 4: Polarization curves for electrodes from DG + 20% CoPc, with different electrolytes: (a)  $10 \text{ g}\cdot\text{L}^{-1}$   $\text{H}_2\text{SO}_3$  +  $18 \text{ g}\cdot\text{L}^{-1}$  NaCl; (b)  $10 \text{ g}\cdot\text{L}^{-1}$   $\text{H}_2\text{SO}_3$  +  $1 \text{ M NaOH}$ ; (c)  $10 \text{ g}\cdot\text{L}^{-1}$   $\text{H}_2\text{SO}_3$  +  $18 \text{ g}\cdot\text{L}^{-1}$  NaCl +  $1 \text{ M NaOH}$ ;  $t^\circ = 25^\circ\text{C}$ .

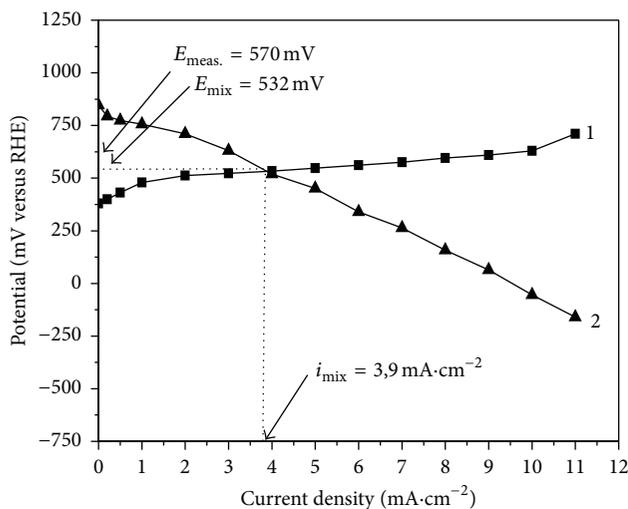
The polarization curves of sulfur dioxide reduction are shown in Figure 3. It can be seen that the substrate used (a) does not have a catalytic effect. The good initial results with electrodes containing DG + 20% CoPc + 6% Pt (d) have not been confirmed by long-term tests due to platinum poisoning. The curves (b) and (c), respectively, DG + 20% CoPc and Perovskite are similar.

The influence of the electrolyte (with or without NaOH and NaCl) on  $\text{SO}_2$  reduction is shown in Figure 4. The figure shows that the absence of NaCl (curve (b)) leads to a conductivity reduction, while NaOH absence leads to a decrease in pH and change in process kinetics, yielding sulfur.

**3.2. Electrochemical Mechanism.** The electrochemical mechanism was studied by juxtaposition of the partial curves of

TABLE 1: Data of initial and final concentration of  $S^{2-}$  and  $SO_3^{2-}$ .

Reagent	Initial amount ( $I_C$ )	Final amount ( $F_C$ )	$\frac{\Delta N}{I_C - F_C}$ per hour	$I = \frac{\Delta N \cdot F}{\Delta t}$ (mA)	$i = I/10$ $\text{mA}\cdot\text{cm}^{-2}$
$C_{SO_2}$ , $\text{mg}\cdot\text{L}^{-1}$	57	37.75	21.25		
$N_{H_2SO_3}$ (gEqv)	$3.56\cdot 10^{-3}$	$2.23\cdot 10^{-3}$	$1.33\cdot 10^{-3}$	35.64	3.56
$C_{S_2^{2-}}$ , $\text{mg}\cdot\text{L}^{-1}$	40	16.5	23.5		
$N_{S^{2-}}$ (gEqv)	$23.5\cdot 10^{-3}$	$9.7\cdot 10^{-3}$	$13.8\cdot 10^{-3}$	37.0	3.7

FIGURE 5: Polarization curves for the oxidation of  $H_2S$  from Figure 2(a) and the reduction of  $SO_2$  from Figure 3(b).

oxidation of hydrogen sulfide (Figure 2(a)) and the reduction of  $SO_2$  (Figure 3(b)) as presented in Figure 5. From the intersection of the curves we can determine the mixed potential and current density, which are  $E = 532\text{ mV}$  and  $i \approx 3.9\text{ mA/cm}^2$ . The overall reaction rate can be estimated using Faraday's equation:

$$I = \frac{\Delta N \cdot F}{\Delta t}, (\text{mA}). \quad (5)$$

The agreement between the mixed potential (in a solution containing hydrogen sulfide and sulfur dioxide  $E_{\text{mix}}(H_2S, SO_2) \approx 570\text{ mV}$  and the potential calculated from the intersection of the partial curves  $E_{\text{mix}}(\text{calculate}) = 532\text{ mV}$ ) is very good, since the deviations for porous electrodes of around  $\Delta E \sim 30\text{ mV}$  are acceptable [33]. This agreement is an indication of the electrochemical mechanism of the process. In order to indubitably prove the electrochemical mechanism we have to measure the overall reaction rate.

**3.3. Estimation of the Overall Reaction Rate in a Flow Type Reactor.** The overall reaction rate has been estimated in a flow type reactor with a solution (electrolyte)  $H_2SO_3 + 18\text{ g}\cdot\text{L}^{-1}\text{ NaCl} + 1\text{ M NaOH} + Na_2S\cdot 9H_2O$ . The electrolyte was circulated all the time. The amount of catalytic mass in the reactor is equal to the catalytic amount in  $10\text{ cm}^2$

immersed electrodes (420 mg 35% teflonized Vulcan XC-72 + 18 mg DG (Standard Degussa carbon black) with 20% CoPc). The inlet and outlet concentrations were found analytically. Unfortunately we do not have a method for instant analysis of the solution in order to find the momentary reaction rate. The overall rate of the oxidation reduction process was estimated analytically after one hour of operation. The whole electrolyte volume of circulating electrolyte is  $V = 200\text{ mL}$ . It can be converted into  $i = \text{mA}$  (electrochemical rate) with the aid of (5). The number of exchanged electrons, following reactions (2) and (3), is  $n = 2$  for  $H_2S$  oxidation and  $n = 4$  for  $SO_2$  reduction. All data are shown in Table 1.

From the good agreement between the analytical and electrochemical results as shown in Table 1, we can conclude that the electrochemical rather than chemical conversion of hydrogen sulfide and sulfur dioxide is preferable.

## 4. Conclusion

The set-up used to accomplish the process discussed can be a continuously stirred tank reactor containing the sea water and the catalyst and bubbles by the stack gases containing sulfur dioxide. The process could be carried out either in acid or alkaline media.

Based on the data in Table 1 we can suppose that the conversion of 18 g hydrogen sulfide per hour into environmentally friendly products like sulfate, sulfur, and polysulfides will require about 460 g of catalyst.

Based on these results we have developed a method for purification of both contaminants at normal atmospheric conditions by electrochemical means. The optimum conditions and suitable catalytic mass have been determined. The realized process completely converts  $H_2S$  and  $SO_2$  into environmentally friendly products (sulfur, polysulfides, etc.). A medium size coal thermal power station of 630 MW releases about 20 metric tons of sulfur dioxide daily which requires  $2\text{ mln m}^3$  (two million cubic meters per hour) of sea water.

One cubic meter of sea water ( $8\text{ mg}\cdot\text{L}^{-1}$ ;  $8\text{ g}\cdot\text{m}^{-3}$ ) cleans three cubic meters of  $SO_2$  containing gases ( $C_{SO_2} = 0.1\% \text{ vol.}$ ;  $\sim 2.85\text{ g}\cdot\text{m}^{-3}$ ).

It seems more realistic to convert reasonable amounts of hydrogen sulfide to produce sulfur and polysulfide as commercial products, thus reducing small amounts of sulfur dioxide, and to remove the rest by traditional methods, like treatment with lime or limestone.

## Conflict of Interests

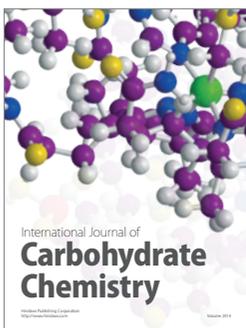
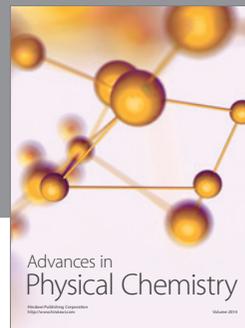
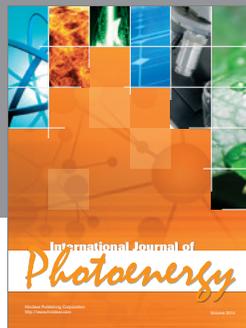
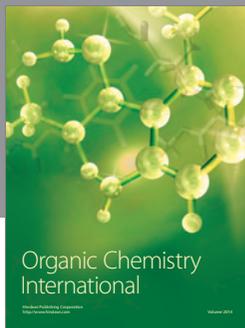
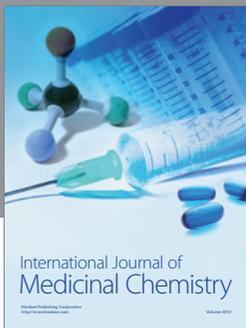
The authors declare that there is no conflict of interests regarding the publication of this paper.

## Acknowledgments

This work was supported by the project Hydrogen Production from Black Sea Water by Sulfide-Driven Fuel Cell HYSULF-CEL (<http://bs-era.net/>) of the 7 FP of the European Union, Contract DNS7RP 01/32, of the Ministry of Education and Science of the Republic of Bulgaria.

## References

- [1] C. F. Claus, British Patent 5958, 1883.
- [2] P. F. Cross, UK Patent Application GB 2503294 A, 2012.
- [3] Z. Nugeired, EA 000764 B1, 1998.
- [4] V. K. Kam-Wang, WO 2001053197 A1 (also US 6214311), 2000.
- [5] M. Rameshni, WO2009055104 A2 (also US 7722852 B2), 2009.
- [6] T. Shimizu, M. Peglow, K. Yamagiwa, and M. Tanaka, "Comparison among attrition-reaction models of SO<sub>2</sub> capture by uncalcined limestone under pressurized fluidized bed combustion conditions," *Chemical Engineering Science*, vol. 58, no. 13, pp. 3053–3057, 2003.
- [7] L. Cholpanova, N. Kazanski, and R. Tsacheva, *Bulgarian Environmental Projects*, Pensoft, Bulgaria, Sofia, 1995.
- [8] K. Petrov, "Electrochemical methods for cleansing of H<sub>2</sub>S and SO<sub>2</sub> in industrial and natural contaminated waters. Elcox method," in *Technological Innovations in Sensing and Detection of Chemical, Biological, Radiological, Nuclear Threats and Ecological Terrorism*, NATO Science for Peace and Security Series A: Chemistry and Biology, pp. 287–299, Springer, Amsterdam, The Netherlands, 2012.
- [9] A. Midilli, M. Ay, A. Kale, and T. N. Veziroglu, "A parametric investigation of hydrogen energy potential based on H<sub>2</sub>S in Black Sea deep waters," *International Journal of Hydrogen Energy*, vol. 32, no. 1, pp. 117–124, 2007.
- [10] T. Chivers, J. B. Hyne, and C. Lau, "The thermal decomposition of hydrogen sulfide over transition metal sulfides," *International Journal of Hydrogen Energy*, vol. 5, no. 5, pp. 499–506, 1980.
- [11] S. Z. Baykara, E. H. Figen, A. Kale, and T. Nejat Veziroglu, "Hydrogen from hydrogen sulphide in Black Sea," *International Journal of Hydrogen Energy*, vol. 32, no. 9, pp. 1246–1250, 2007.
- [12] K. Petrov and S. Srinivasan, "Low temperature removal of hydrogen sulfide from sour gas and its utilization for hydrogen and sulfur production," *International Journal of Hydrogen Energy*, vol. 21, no. 3, pp. 163–169, 1996.
- [13] L. Goltseva and A. Yutkin, "Black sea," *Inventor*, vol. 2, p. 7, 1996.
- [14] K. Petrov, S. Z. Baykara, D. Ebrasu, M. Gulin, and A. Veziroglu, "An assessment of electrolytic hydrogen production from H<sub>2</sub>S in Black Sea waters," *International Journal of Hydrogen Energy*, vol. 36, no. 15, pp. 8936–8942, 2011.
- [15] I. Streeter, A. J. Wain, J. Davis, and R. G. Compton, "Cathodic reduction of bisulfite and sulfur dioxide in aqueous solutions on copper electrodes: an electrochemical ESR study," *Journal of Physical Chemistry B*, vol. 109, no. 39, pp. 18500–18506, 2005.
- [16] Z. Mao, A. Anani, R. E. White, S. Srinivasan, and A. J. Appleby, "Modified electrochemical process for the decomposition of hydrogen sulfide in an aqueous alkaline solution," *Journal of the Electrochemical Society*, vol. 138, no. 5, pp. 1299–1303, 1991.
- [17] J. Zaman and A. Chakma, "Production of hydrogen and sulfur from hydrogen sulfide," *Fuel Processing Technology*, vol. 41, no. 2, pp. 159–198, 1995.
- [18] K. Petrov, I. Nikolov, T. Vitanov, D. Uzun, and V. Ognjanov, "Pyrolyzed Co-phtalocyanine as a catalyst for the oxidation of sulphur dioxide," *Bulgarian Chemical Communications*, vol. 42, no. 3, pp. 189–193, 2010.
- [19] K. T. Chuang, J. C. Donini, A. R. Sanger, and S. V. Slavov, "A proton—conducting solid state H<sub>2</sub>S-O<sub>2</sub> fuel cell 2. Production of liquid sulphur at 120–145°C," *International Journal Hydrogen Energy*, vol. 25, no. 9, pp. 887–894, 2000.
- [20] C. Wagner and W. Z. Traud, "Über die deutung von korrosionsvorgängen durch überlagerung von elektrochemischen teilvorgängen und über die potentialbildung an mischelektroden," *Zeitschrift für Elektrochemie und Angewandte Physikalische Chemie*, vol. 44, pp. 391–402, 1938.
- [21] V. V. Scorcelletti, *Theoretical Fundamentals of Corrosion in Metals*, Khimia Leningrad: Chemistry, Saint Petersburg, Russia, 1973.
- [22] M. Spiro and P. L. Freund, "Polyelectrodes: evidence for and against the wagner and traud additivity hypothesis," *Journal of Electroanalytical Chemistry*, vol. 144, no. 1-2, pp. 293–301, 1983.
- [23] K. Vetter, *Electrochemical Kinetics*, Chemistry, Moscow, Russia, 1967.
- [24] B. V. Romanovsky, *Fundamentals of Chemical Kinetics*, Exam, Moscow, Russia, 2006.
- [25] D. Uzun, E. Razkazova-Velkova, V. Beschkov, and K. Petrov, "The electrochemical method for separation of sulphur dioxide and hydrogen sulphide from fluids," Bulgarian Patent Office, Patent Request Number: 111701, 2014.
- [26] D. Uzun, P. Hiev, D. Vladikova et al., "Electrocatalytic oxidation-reduction reactions of metal-hydride alloys with teflon-carbon additives," *Bulgarian Chemical Communications*, vol. 42, no. 2, pp. 113–118, 2010.
- [27] N. P. Penchev and B. N. Zagorghev, *Analytic Chemistry, Quantity Analysis*, Science and Art, Sofia, Bulgaria, 1956.
- [28] A. Busev and L. Simon, *Analytical Chemistry of Sulphur*, Science, Moscow, Russia, 1975.
- [29] W. Fresenius and G. Jander, *Handbook for Analytical Chemistry*, Springer, 4th edition, 1967.
- [30] N. P. Penchev and B. N. Zagorghev, *Analytic Chemistry Qualitative Analysis*, Science and Art, Sofia, Bulgaria, 1964.
- [31] M. G. Bapat and B. Sharma, "Application of amperometric dead stop end point to iodate procedures in presence of mercuric mercury," *Fresenius' Zeitschrift für Analytische Chemie*, vol. 157, no. 4, pp. 258–261, 1957.
- [32] D. Uzun, E. Razkazova-Velkova, K. Petrov, and V. Beschkov, "Electrochemical method for energy production from hydrogen sulfide in the Black sea waters in sulphide—driven fuel cell," *Bulgarian Chemical Communications*, vol. 47, no. 3, pp. 859–866, 2015.
- [33] A. Carbó, *Electrochemistry of Porous Materials*, Taylor & Francis Group, 2012.



**Hindawi**

Submit your manuscripts at  
<http://www.hindawi.com>

