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Energy exploitation of acid gas with high H₂S content by means of a chemical looping combustion system

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Abstract

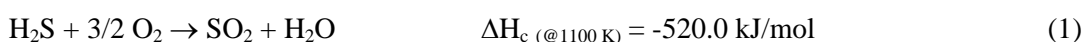
In gas and petroleum industry, the waste gas stream from the sweetening process of a sour natural gas stream is commonly referred as acid gas. Chemical Looping Combustion (CLC) technology has the potential to exploit the combustible fraction of acid gas, H₂S, to produce energy obtaining a flue gas highly concentrated on CO₂ and SO₂, which can be cost-effectively separated for subsequent applications, such as sulfuric acid production. At the same time, a concentrated CO₂ stream ready for storage is obtained. The resistance of oxygen carriers to sulfur becomes crucial when an acid gas is subjected to a CLC process since the H₂S content can be very high. In this work, a total of 41 hours of continuous operation with acid gas and H₂S concentrations up to 20 vol.% has been carried out in a 500 W_{th} CLC unit with two oxygen carriers based on Cu (Cu₁₄γAl) and Fe (Fe₂₀γAl). The formation of copper sulfides and the SO₂ emissions in the air reactor made the Cu₁₄γAl material not adequate for the process. In contrast, excellent results were obtained during acid gas combustion with the Fe₂₀γAl oxygen carrier.

H₂S was fully burnt to SO₂ in the fuel reactor at all operating conditions, SO₂ was never detected in the gas outlet stream of the air reactor, and iron sulfides were never formed even at H₂S concentrations as high as 20 vol.%. Furthermore, it was found that a H₂S content of 20 vol.% in the acid gas was high enough to turn the CLC process into an auto-thermal process. Based on these results, it can be concluded that the Fe-based materials prepared by impregnation are very adequate to exploit the energy potential of acid gas mixtures with CO₂ capture.

Keywords. Acid gas, Chemical Looping Combustion, H₂S, H₂SO₄, oxygen carrier.

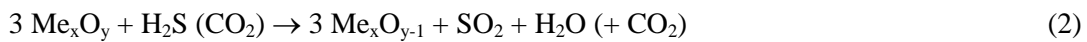
1. Introduction

In gas and petroleum industry, the waste gas stream from the sweetening process of a sour natural gas stream is commonly referred as acid gas, which contains significant amounts of H₂S and CO₂. Usually, this gas is subsequently sent to a Sulfur Recovery Unit (SRU). The core part of the SRU is the Claus plant, in which the H₂S is converted into elemental sulfur. The gaseous effluent from the Claus plant still remains 2-5 wt. % of the total sulfur, which must be further processed in a TGU (Tail Gas Unit) to meet emission standards for SO₂. This technology allows the gas and petroleum industry to recover as much as 99.9 wt. % of the total sulfur separated from natural gas [1]. Different alternatives to deal with acid gas mixtures have emerged in the last years, such as the injection of compressed acid gas into porous formations, or its solubilization into light hydrocarbons solvent and subsequent injection into depleted reservoirs as a miscible flood Enhanced Oil Recovery (EOR) technique [2-3]. However, these technologies do not exploit the energy potential of its combustible fraction, i.e. H₂S. It must be considered that the oxidation of H₂S is clearly exothermic, as shown in reaction (1).



Within the options based on the energy exploitation of the acid gas, Chemical-Looping Combustion (CLC) emerges as a possible technology. This combustion process is based on the transfer of the oxygen from air to the fuel by means of a solid oxygen carrier avoiding direct contact between fuel and air. CLC has the advantage of an inherent CO₂ separation, avoiding additional separation units and energy penalties. Different gaseous, solids and liquid fossil fuels have been successfully tested in CLC units during the last years [4-8]. However, CLC technology is also open to new other fuels. Figure 1 shows the technology train to deal with acid gases based on the energy exploitation of this type of gaseous fuel through CLC.

In CLC, the oxygen carrier circulates between two interconnected fluidized beds, the fuel reactor (FR) and the air reactor (AR). In the case of acid gas combustion, H₂S would be oxidized in a first step to SO₂ and H₂O in the FR by a metal oxide which is reduced to a metal or to a reduced form of the corresponding metal oxide (reaction 2). In a second step, the metal or reduced metal oxide is oxidized with air in the AR regenerating the material for a new cycle (reaction 3). After H₂O condensation, a stream only composed by CO₂ and SO₂ would be obtained. In a next step, CO₂ can be easily separated from SO₂ leading to a pure CO₂ stream, ready for compression, transport and storage. On other hand, the SO₂ pure stream can be used for subsequent applications such as the production of sulfuric acid.



Shell Global Solutions International BV company has developed a patent of an improved process for producing H₂SO₄ with CO₂ capture [9]. In this process, CLC of acid gas with subsequent capture of SO₂ and conversion to H₂SO₄ is used. In this patent, it is commented that the plant economics of this new process in comparison with other processes to produce sulfuric acid is significantly more attractive due to the use of smaller, and hence, cheaper equipment and the lesser energy requirements. In addition, any amount of CO₂ produced during combustion of the acid gas by CLC can be recovered efficiently.

The sulfur fed to a CLC plant can be released by different streams. If SO_2 is produced in the FR gas stream will affect to the quality of the CO_2 obtained. At this moment, there are no regulations about the SO_2 concentration allowed for CO_2 storage since this is still a technology under development [10-11]. Guidance values of concentration for the different gases that are usually present in a CO_2 stream ready for its transport and storage can be found elsewhere [12]. Furthermore, although it is not desirable, sulfur can be also released as SO_2 in the AR gas stream, and the corresponding air emission standards and regulations must be fulfilled. Finally, the H_2S can also react with the oxygen carriers to form metal sulfides. The poisoning of the material can be reflected in terms of a significant drop of its combustion efficiency, reactivity and oxygen transport capacity.

Therefore, the resistance of oxygen carriers to the presence of sulfur in fuels becomes crucial when an acid gas is subjected to a CLC process since the amount of H_2S in an acid gas mixture can be very high, as high as 40 vol.% [2]. Our research group at Instituto de Carboquímica (ICB-CSIC) has studied the performance of four highly reactive oxygen carriers in a 500 W_{th} continuous CLC unit when the fuel contained variable amounts of H_2S (up to 3400 vppm) [13-16]. These oxygen carriers were based on Ni, Cu, Fe and Mn oxides respectively. The best results in terms of fuel combustion performance, reactivity or material deactivation were achieved with the Cu- and Fe-based oxygen carriers [14-15]. These materials were further tested for the combustion of sour gas with very high H_2S content, up to 15 vol.% H_2S [17]. The objective was to demonstrate the possibility to use this fuel for thermal energy conversion without the need of a sweetening pre-treatment with improved capture of the CO_2 and SO_2 produced.

In this paper, the aforementioned Cu- and Fe-based oxygen carriers were assessed for acid gas combustion in a continuous 500 W_{th} CLC prototype using H_2S concentrations up to 20 vol.%. The influence of H_2S concentration on the gas products distribution, sulfur splitting between FR and AR and oxygen carrier deactivation has been investigated. Furthermore, an analysis of the possible energy exploitation of the acid gas was carried out.

2. Experimental

Two synthetic oxygen carriers prepared by impregnation on $\gamma\text{-Al}_2\text{O}_3$ as support have been used in this work: one based on copper and another one based on iron. These materials were designated as Cu14 γ Al and Fe20 γ Al oxygen carriers. The method of preparation and the main physical and chemical properties of both materials were deeply explained elsewhere [18-19].

Acid gas combustion tests were carried out in the ICB-CSIC-g1 facility (500 W_{th}) modified for the safe use of high H_2S content fuels. A detailed description of the installation as well as of the gas analysis system can be found elsewhere [17]. A total of 41 hours of continuous operation with acid gas and different H_2S concentrations up to 20 vol. % were carried out, from which 23 hours corresponded to the Cu-based oxygen carrier and 18 hours to the Fe-based material. It must be remarked that the acid gas combustion tests with the Fe-based material were performed with the same batch of particles previously used during 20 h for the sour gas combustion [17]. Tables 1 and 2 shows a summary of the different operating conditions used during the acid combustion tests. The oxygen carrier-to-fuel ratio, ϕ , was defined as the ratio between the oxygen supplied by the oxygen carrier and the oxygen needed to stoichiometrically react with the fuel flow. This parameter has been calculated considering the solids circulation rate, f_s , used in each test. A value of $\phi = 1$ corresponds to the stoichiometric MeO amount needed for complete conversion of the acid gas to SO_2 and H_2O . It must be remarked test Fe4 was carried out with a H_2S concentration of 20 vol.%. This is the highest H_2S concentration ever tested in a CLC unit.

3. Results

3.1 Tests of acid gas combustion with the Cu14- γ Al oxygen carrier

A series of experimental tests were performed in the continuous CLC unit in order to assess the performance of the Cu14- γ Al material for acid gas combustion. Reactivity, combustion efficiency, fluidization behavior, and sulfur splitting between reactors were analyzed.

As an example, Figure 2 shows the results obtained in test Cu3. This Figure illustrates the gas composition obtained at the outlet of the FR and AR and the instantaneous sulfur mass balance. This mass balance, expressed as mol of sulfur per hour, includes the sulfur fed to the system, the sulfur detected at the outlet of the FR and AR, and the total sulfur in the gas phase obtained from both streams. It was observed that H₂S was fully burnt since this compound was never detected in the FR gas outlet stream. The sulfur was mainly transformed into SO₂ in the FR. In addition, variable amounts of SO₂ were detected at the outlet of the AR, reaching values ranged from 200 to 400 vppm (572-1144 mg/Nm³). If these values were normalized to an excess of 6 vol.% O₂, the SO₂ emissions were higher than 200 mg/Nm³, which is the EU emissions limit for boilers higher than 200 MW_{th}.

The appearance of SO₂ in the outlet stream from the AR would indicate the formation of Cu₂S in the FR and its subsequent decomposition in the AR through reactions (4) and (5) considering CuO and CuAl₂O₄ as active copper-based species present in the oxygen carrier particles.



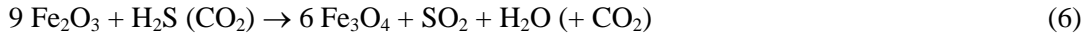
The presence of sulfur in the oxygen carrier particles after operation was corroborated by SEM-EDX analysis and by elemental analysis in a Thermo Flash 1112 elemental analyzer. These results are similar to the ones found during sour gas combustion with this material [17]. Therefore, the utilization of Cu-based oxygen carriers in a CLC process using acid gas as fuel is not recommended.

3.2 Tests of acid gas combustion with the Fe₂₀Al oxygen carrier

3.2.1 Thermodynamic calculations for iron materials

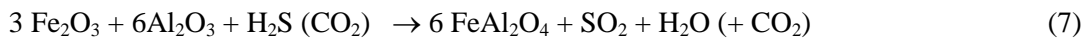
A thermodynamic study was carried out using the HSC Chemistry 6.1 [20] software to determine the fate of sulfur when Fe-based oxygen carriers are used in a CLC plant with acid gas. This program obtains the equilibrium composition by using the method of minimization of the Gibbs free energy of the system for a fixed mass balance, at constant pressure and

temperature. The calculations were conducted considering acid gas as fuel with H₂S concentrations ranged from 1 to 30 vol.%, and operating temperatures from 1073 K to 1273 K. Figure 3a shows the thermodynamic equilibrium of the compounds existing in the FR as a function of the parameter ϕ when the redox pair considered is Fe₂O₃-Fe₃O₄ (see equation 6). The temperature and the H₂S concentration selected for this example were 1223 K and 15 vol.%, respectively.



The thermodynamics for the reaction of acid gas with iron compounds shows a very promising behavior since SO₂ is the unique sulfur compound possible at ϕ values even below 1, in this case for $\phi > 0.75$. At lower values of ϕ , several sulfur compounds appear, being possible the formation of several iron sulfides (FeS and Fe_{0.88}S) together with other gas sulfur compounds (SO₂, H₂S, COS) and even elemental sulfur (S₂).

However, in Fe-based oxygen carriers prepared by impregnation on alumina, FeAl₂O₄ can be formed as reduced compound [15, 19, 21]. Therefore, new thermodynamic calculations including Al₂O₃ as solid phase were carried out, and the results are shown in Figure 3b. In this case, the value of $\phi = 1$ corresponds to the stoichiometric amount of Fe₂O₃ (Al₂O₃) needed for a full conversion of the fuel, H₂S, to SO₂ and H₂O when the redox pair considered is Fe₂O₃ (Al₂O₃) - FeAl₂O₄.



As in the previous calculations, at ϕ values higher than 1, the unique sulfur specie was SO₂. However, interesting results were obtained at substoichiometric conditions. In this case, iron sulfides formation was only possible at very low values of ϕ ($\phi < 0.2$). In most of the substoichiometric region of ϕ , ($0.1 < \phi < 1$) the unique stable iron specie is FeAl₂O₄ because the presence of H₂O avoided the presence of elemental Fe, which is the responsible for the iron sulfides formation. In this zone, most of the sulfur compounds are in the gas phase as H₂S, SO₂, and S₂. The lower concentrations of CO and H₂ present in the acid gas environment compared to the existing ones in a sour gas environment [17] represent an advantage with respect to the

possible formation of iron sulfides. The absence of these compounds at very low ϕ values during acid gas combustion can be considered as a remarkable result, and even more if it is taken into consideration that the transformation $\text{Fe}_2\text{O}_3\text{-FeAl}_2\text{O}_4$ supposes to increase 3 times the oxygen transport capacity with respect to the use of the pair $\text{Fe}_2\text{O}_3\text{-Fe}_3\text{O}_4$ reaching complete fuel combustion [15, 19, 22-23]. Thermodynamic calculations made at different temperatures from 1073 to 1273 K showed similar results.

3.2.2 Combustion tests in the ICB-CSIC-g1 facility

A total of 18 hours of continuous operation was carried out with the $\text{Fe}_{20}\gamma\text{Al}$ oxygen carrier for the combustion of acid gas. Reactivity, combustion efficiency, fluidization behavior, and sulfur splitting between reactors were analyzed.

Figure 4 shows the gas composition obtained at the outlet of the FR and AR for tests Fe3 and Fe4, which are the tests with the highest H_2S feeding concentrations, i.e., 15 and 20 vol. % H_2S . Moreover, a graph showing the corresponding sulfur mass balances was added. In all operating conditions the H_2S was fully burnt to SO_2 in the FR, and no SO_2 was detected in the outlet gas stream of the AR at any time and operating condition. It must be remarked that high H_2S concentrations, up to 20 vol.%, were used. The results obtained were very satisfactory, and even better than during sour gas combustion [17]. This is due to the higher oxygen carrier-to-fuel ratios used during the acid gas tests and the more favorable thermodynamic properties of the Fe-based materials for acid gas combustion regarding iron sulfides formation.

With the above experimental data, a sulfur mass balance was performed for the different tests. Figure 5 shows this balance considering the sulfur released as SO_2 in both the FR and the AR. It can be observed that sulfur only appeared in the FR. The mass balance closed with less than 10 % error in all tests. According to the thermodynamic analysis showed in Section 3.2.1, no iron sulfides should be formed during operation taking into consideration the high oxygen carrier-to-fuel ratios used along the experimental campaign. However, a deeply characterization of the solid samples extracted from the system was carried out to confirm it.

3.2.3 Characterization of the oxygen carrier

Different samples of material extracted from the FR and AR during operation were used for solid characterization. The crystalline phases present in the oxygen carrier particles were determined from powder XRD patterns. α -Al₂O₃ and FeAl₂O₄ were the main phases found in the FR material, and α -Al₂O₃ and Fe₂O₃ were the corresponding ones in the AR.

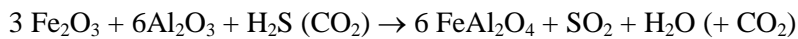
In addition, elemental analysis showed that sulfur was not present in any of the samples. Also, a SEM-EDX analysis was performed to oxygen carrier particles extracted from the CLC unit after the different tests. As, an example Figure 6 shows the SEM-EDX image of a cross-section of a particle after test Fe4, the experiment with the highest H₂S concentration used, 20 vol.%. This analysis confirmed again the absence of sulfur inside the particles. Consequently, it can be stated that the deviations in the sulfur mass balance showed in Figure 5 should be due to experimental errors.

Finally, TGA characterization demonstrated that used oxygen carrier particles maintained a high reactivity after 18 hours of operating time in the CLC unit even considering the high H₂S content present in the acid gas.

4. Energy exploitation of acid gas

The energy exploitation of an acid gas stream in a CLC unit, and the corresponding thermal integration of the whole CLC system, depends on different factors such as the use of a specific oxygen carrier, the solids recirculation rate (related with the oxygen carrier-to-fuel ratio, ϕ) and the energy potential of the fuel in terms of the H₂S content present in the feeding stream.

According to the previous experimental results, only the Fe₂₀Al material has been demonstrated to be adequate for the combustion of acid gas with high H₂S concentrations without any SO₂ emissions to the atmosphere. Therefore, the analysis of the possible energy exploitation of the acid gas has been carried out with this material. The reactions in the FR and AR would be the following,



$$\Delta H_{\text{r}} (@_{1100 \text{ K}}) = -44.7 \text{ kJ/mol} \quad (8)$$



The energy and mass balances in a CLC process are related to the reaction heat and the oxygen transport capacity of the oxygen carrier. The reduction reaction can be exothermic or endothermic depending on the redox system considered. In this case, the reduction reaction of Fe_2O_3 (Al_2O_3) with H_2S to give FeAl_2O_4 is exothermic, see reaction (8). This fact means that the solids circulation rate in a CLC unit for acid gas combustion is not fully limited by the heat balance.

The solids recirculation rate, m_{OC} , depends on the rate of oxygen transfer needed for complete combustion of the acid gas to SO_2 and H_2O (see reaction 8), m_o , on the oxygen ratio of the redox pair considered, R_o , on the metal oxide content of the oxygen carrier, x_{OC} , and on the variation of solids conversion in the reactors, ΔX_s ,

$$m_{OC} = \frac{\dot{m}_o}{R_o \cdot x_{OC} \cdot \Delta X_s} \quad (10)$$

To obtain 1 MW of power from the use of H_2S as fuel, the transport of 5.8 mol O/s is necessary (m_o), similar to the 5 and 3.8 mol O/s corresponding to the combustion of natural gas and syngas, respectively. This result proves the high energy potential of the acid gas. R_o is the mass fraction of oxygen that can be used in the oxygen transfer, being 0.1 for the pair $\text{Fe}_2\text{O}_3/\text{FeAl}_2\text{O}_4$. The metal oxide content, x_{OC} , in the $\text{Fe}_{20}\gamma\text{Al}$ oxygen carrier was 20 wt.%. Finally, ΔX_s is directly related to the oxygen carrier to fuel ratio, ϕ . Assuming a variation of solids conversion of $\Delta X_s = 1$, the solids recirculation rate, m_{OC} , per MW of H_2S is 4.6 kg/s. These values are within the usual operating range in a CLC process [24] and would not be a limiting factor.

Finally, as above mentioned, the H_2S content in an acid gas mixture can vary in a great extent. Therefore, an analysis of the use of the energy potential of acid gas in a CLC process as a function of the H_2S content was carried out. Figure 7 shows the scheme of the CLC process considered. The assumptions and parameters used for this study were the following: the fuel reactor was adiabatic and full combustion of the acid gas was achieved, the temperature in the

air reactor was fixed at 1223 K, the oxygen carrier-to-fuel ratio (ϕ) took a value of 2, an air excess of 20 vol.% was used in the air reactor ($\lambda = 1.2$), the temperature of the inlet gas streams was 298 K, and the temperatures of the outlet gas streams were the same to the existing ones in the reactors. A 1 MW_{th} of acid gas power was taken as reference in all cases.

The temperature in the FR was given by a balance among the heat generated in the exothermic reaction between the H₂S and the oxygen carrier (reaction 8), the heat contained in the hot solids coming from the AR, and the heat needed to raise the temperature of the acid gas stream. Moreover, the heat extracted from the AR depended on the heat generated in the exothermic oxidation reaction (reaction 9), the heat needed to raise the temperature of the air stream, and on the heat transported by the solids coming from the FR.

Figure 8 shows the thermal power that could be removed from the AR and the temperature of the FR as a function of the volumetric percentage of H₂S in the acid gas stream fed to a CLC unit. From this Figure, it can be concluded that a H₂S content of 20 vol.% in the acid gas was high enough to turn CLC into an auto-thermal process. This result means that the heat obtained from the exothermic reaction between the Fe-based oxygen carrier particles and H₂S compensates the heat absorbed by the amount of CO₂ present in the acid gas stream to reach a temperature of 1198 K, only 25 K lower than the temperature fixed at the AR. This is an outstanding result since the Fe₂₀γAl material had already demonstrated an excellent performance in terms of stability, reactivity and combustion behavior at these conditions. Furthermore, if the H₂S content was higher than 75 vol.%, the temperature of the FR would be higher than the 1223 K existing in the AR, and almost 30 % of the energy potential of the acid gas could be extracted from the AR to use it in additional processes. Considering a typical acid gas composition with a 35 vol.% H₂S content [9], the temperature in the fuel reactor would be 1208 K and 18 % of the energy potential of the acid gas could be used for further energy applications.

5. Conclusions

In this work, two oxygen carriers based on copper and iron respectively were subjected to acid gas combustion tests in a continuous 500 W_{th} CLC unit with high H₂S concentrations. The Cu₁₄γAl oxygen carrier was able to burn completely the acid gas to SO₂, being mainly released in the FR. However, some SO₂ amounts were also detected in the outlet stream from the AR, which usually exceeded the EU emission limits established for industrial boilers. In addition, some sulfur, possibly as Cu₂S, was accumulated in the oxygen carrier particles during operation at all operating conditions. According to these results, Cu-based oxygen carriers cannot be proposed to exploit the energy potential of acid gas in a CLC process during long operation times.

In contrast, very satisfactory results were obtained with the Fe₂₀γAl oxygen carrier. This material was always able to burn completely the acid gas, SO₂ was never detected in the gas outlet stream of the AR, and iron sulfides were not formed at any experimental condition.

Furthermore, the analysis of the energy exploitation of the acid gas through CLC technology showed that a H₂S content of 20 vol.% was high enough to turn CLC into an auto-thermal process. This is a remarkable result considering that the Fe₂₀γAl material demonstrated an excellent performance in terms of stability, reactivity and combustion behavior at these conditions. Therefore, it can be concluded that Fe-based materials prepared by impregnation are very adequate to exploit the energy potential of acid gas streams without any SO₂ emission into the atmosphere and with an efficient recovery of the CO₂.

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Figure Captions

Figure 1. Scheme to deal with acid gases through CLC technology.

Figure 2. Gas composition at the outlet stream from the FR and AR, and sulfur balance corresponding to test Cu3 with the Cu14 γ Al material.

Figure 3. Thermodynamic calculations for the systems (a) Fe₂O₃-Fe₃O₄/acid gas, and (b) Fe₂O₃(Al₂O₃)-FeAl₂O₄/acid gas. Acid gas composition: 15 vol.% H₂S-85 vol.% CO₂. T = 1223K.

Figure 4. Gas composition at the outlet stream from the FR and AR, and sulfur balance corresponding to tests Fe3 and Fe4 with the Fe20 γ Al oxygen carrier.

Figure 5. Sulfur mass balance for the combustion of acid gas with the Fe-based oxygen carrier.

Figure 6. SEM-EDX image of a cross-section of a Fe20 γ Al oxygen carrier particle after test Fe4 in the CLC unit.

Figure 7. Scheme of the CLC process considered to analyse the energy potential of acid gas.

Figure 8. Relation between the H₂S content present in an acid gas stream (vol.%) and the thermal power extracted from the AR, and the temperature of the FR. T_{AR} = 1223 K; $\phi = 2$; Power = 1 MW_{th} of acid gas; $\lambda=1.2$.

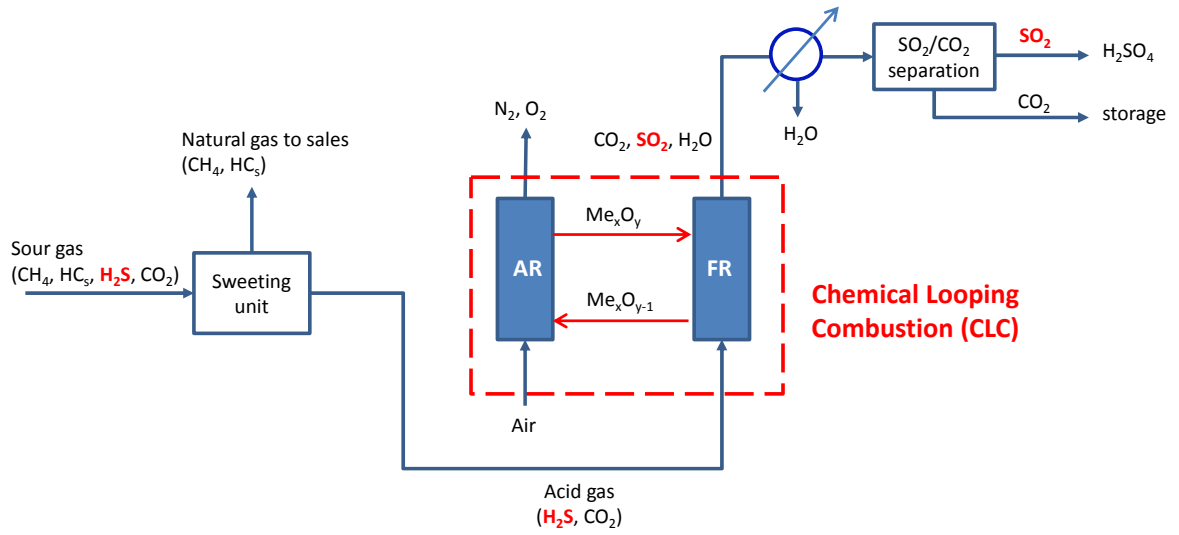


Figure 1. Scheme to deal with acid gases through CLC technology.

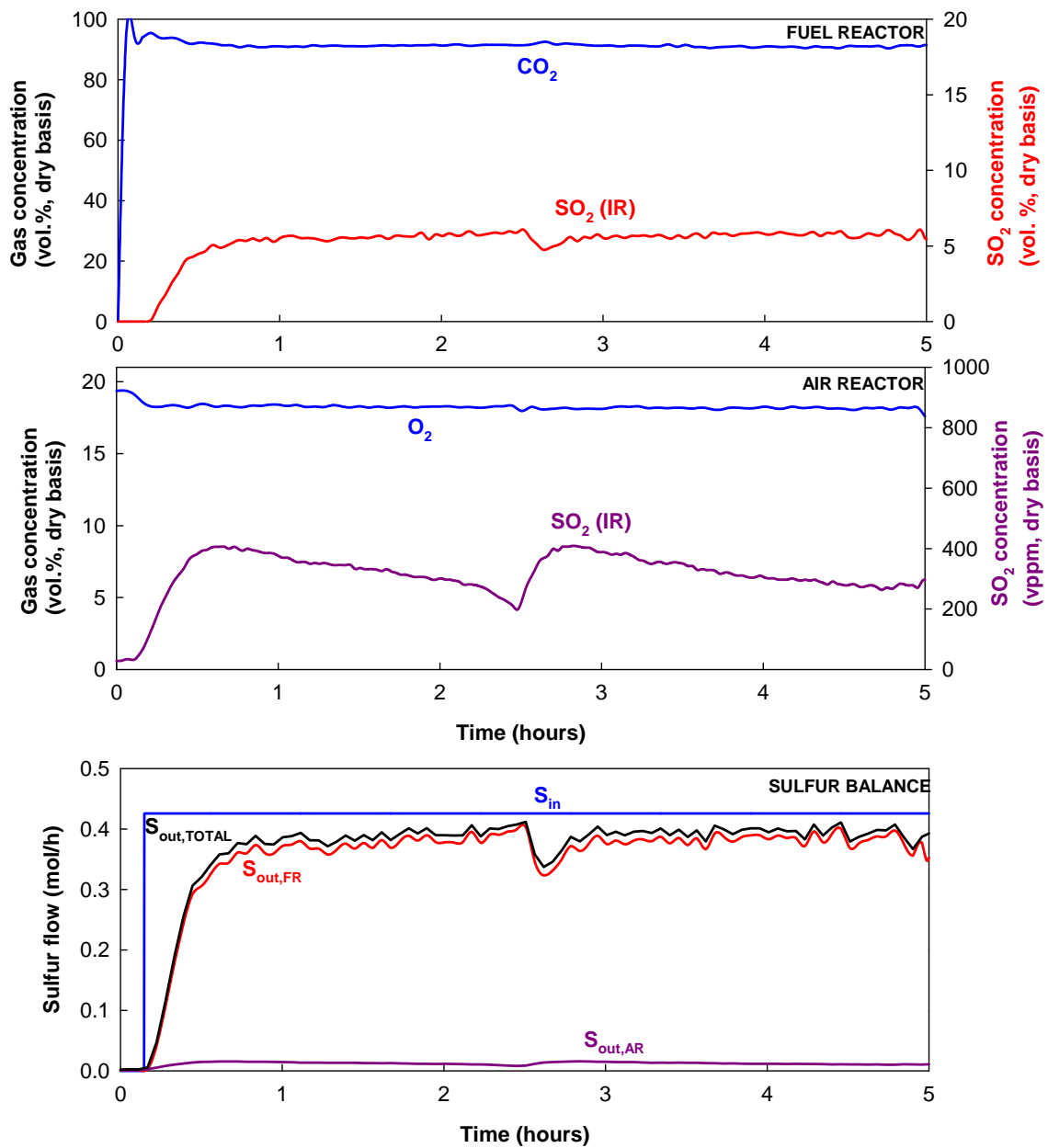


Figure 2. Gas composition at the outlet stream from the FR and AR, and sulfur balance corresponding to test Cu3 with the Cu14 γ Al material.

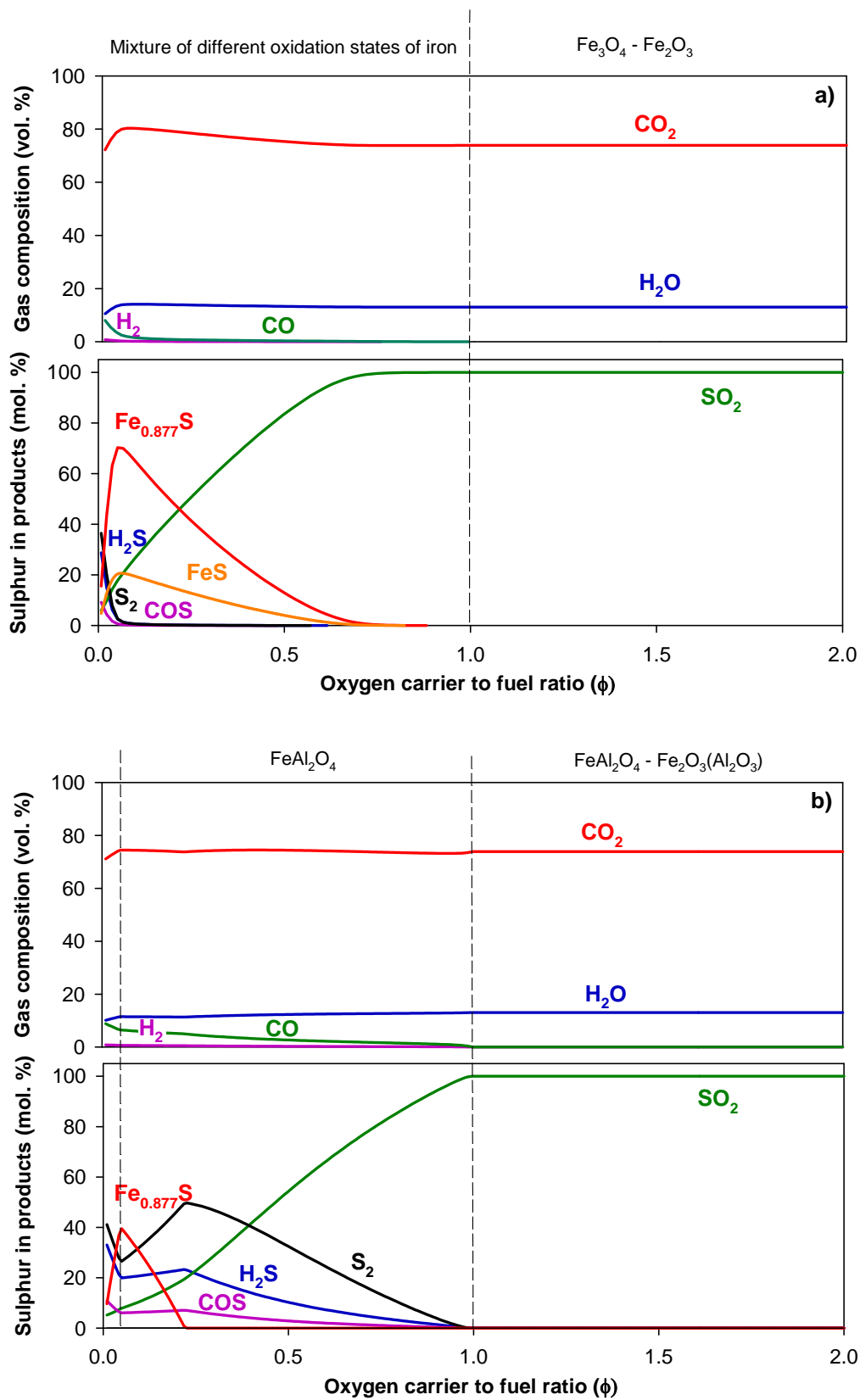


Figure 3. Thermodynamic calculations for the systems (a) $\text{Fe}_2\text{O}_3\text{-Fe}_3\text{O}_4/\text{acid gas}$, and (b) $\text{Fe}_2\text{O}_3(\text{Al}_2\text{O}_3)\text{-FeAl}_2\text{O}_4/\text{acid gas}$. Acid gas composition: 15 vol.% H_2S -85 vol.% CO_2 . $T = 1223\text{K}$.

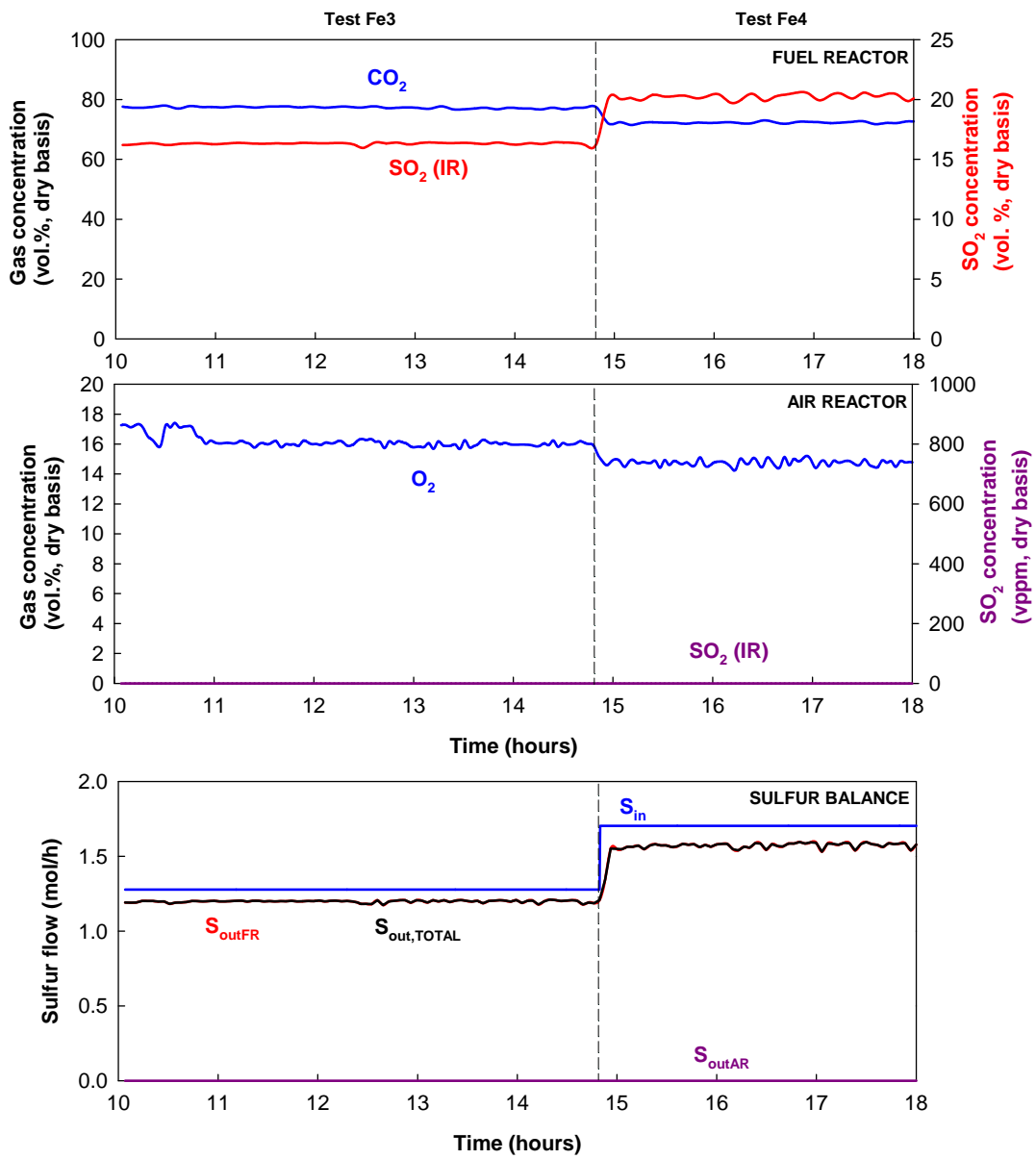


Figure 4. Gas composition at the outlet stream from the FR and AR, and sulfur balance corresponding to tests Fe3 and Fe4 with the Fe₂₀γAl oxygen carrier.

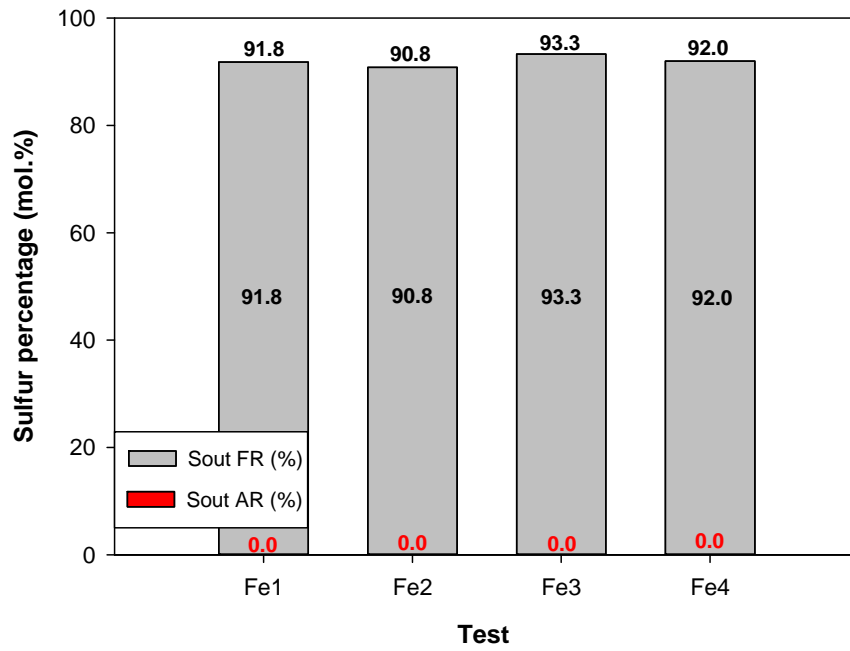


Figure 5. Sulfur mass balance for the combustion of acid gas with the Fe-based oxygen carrier.

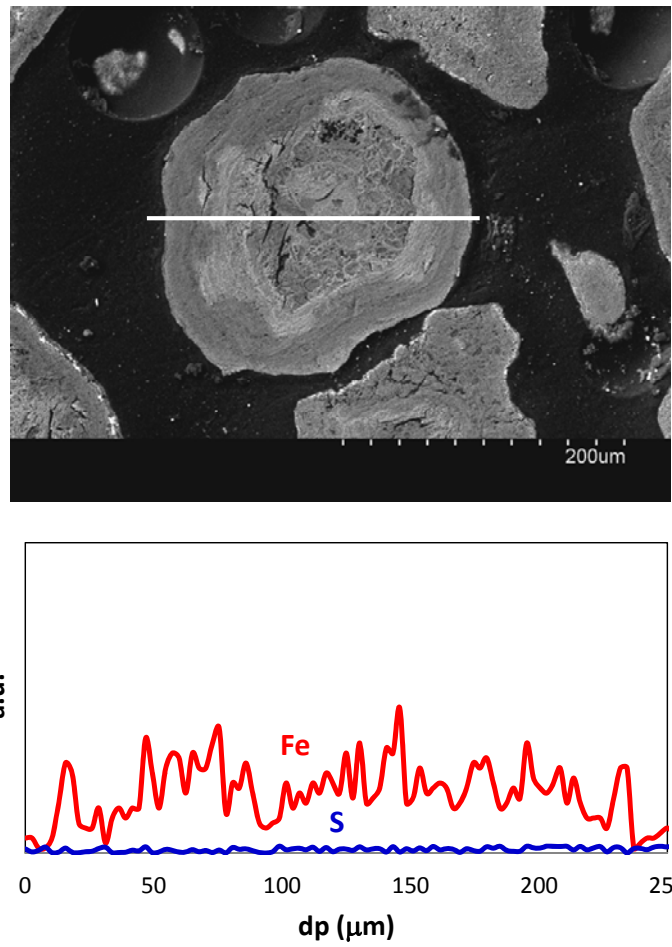


Figure 6. SEM-EDX image of a cross-section of a $\text{Fe}_{20}\gamma\text{Al}$ oxygen carrier particle after test Fe4 in the CLC unit.

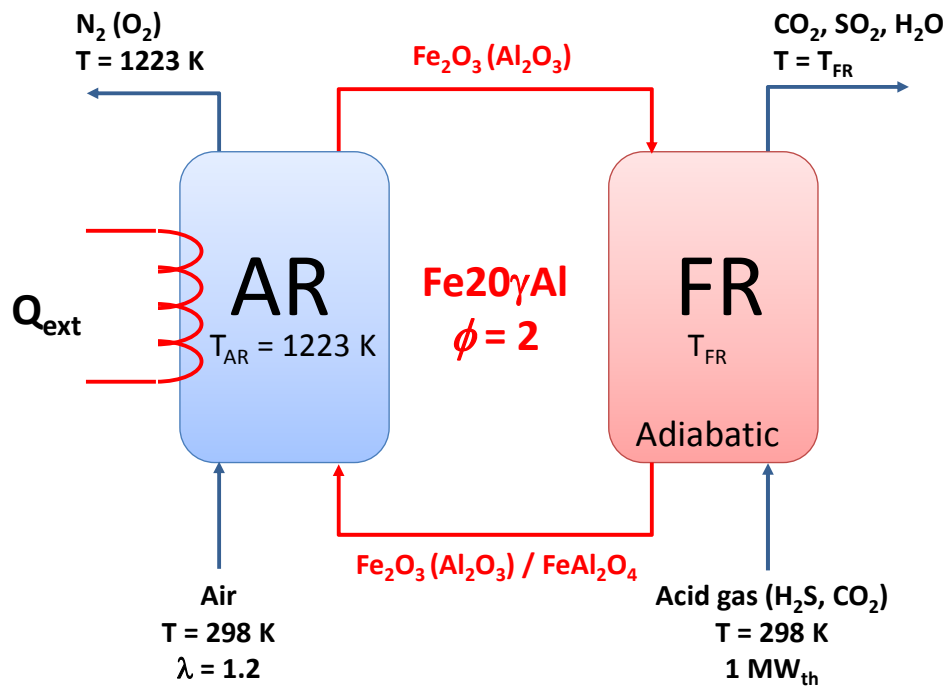


Figure 7. Scheme of the CLC process considered to analyze the energy potential of acid gas.

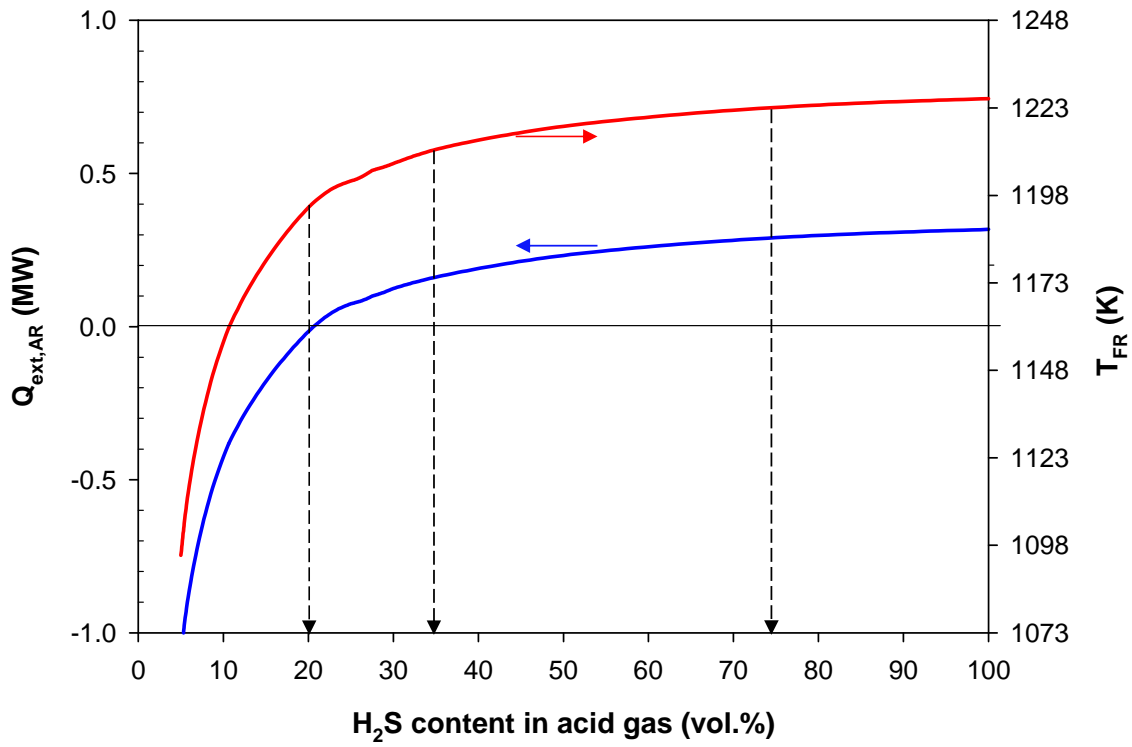


Figure 8. Relation between the H₂S content present in an acid gas stream (vol.%) and the thermal power extracted from the AR, and the temperature of the FR. T_{AR} = 1223 K; $\phi = 2$; Power = 1 MW_{th} of acid gas; $\lambda=1.2$.

Tables

Table 1. Combustion tests of acid gas with the Cu-based oxygen carrier.

Table 2. Combustion tests of acid gas with the Fe-based oxygen carrier.

Table 1. Combustion tests of acid gas with the Cu-based oxygen carrier.

Test	T _{FR}	T _{AR}	Gas composition (vol.%)				f _s ^a	ϕ ^b	Cumulative time
			CO ₂	H ₂	H ₂ O	H ₂ S			
	(K)	(K)					(kg/h)	(h)	
Cu1	1073	1123	80	5	10	0	10.9	42.8	0.5
Cu2	1073	1123	80	5	10	5	10.9	10.7	1.0
Cu3	1073	1123	70	5	20	5	10.9	10.7	6.0
Cu4	1073	1123	65	5	20	10	10.9	6.1	12.0
Cu5	1073	1123	60	5	20	15	10.3	4.0	17.0
Cu6	1073	1123	75	5	20	0	11.8	46.3	17.5
Cu7	1073	1123	70	5	20	5	11.8	11.6	18.5
Cu8	1073	1123	65	5	20	15	11.8	4.6	21.0
Cu9	1073	1123	75	5	20	0	11.8	4.3	23.0

^a f_s = solids circulation rate; ^b ϕ = oxygen carrier-to-fuel ratio

Table 2. Combustion tests of acid gas with the Fe-based oxygen carrier.

Test	T _{FR} (K)	T _{AR} (K)	Gas composition (vol.%)				f _s ^a (kg/h)	ϕ ^b	Cumulative time (h)
			CO ₂	H ₂	H ₂ O	H ₂ S			
Fe1	1173	1223	70	5	20	5	13.1	9.3	5
Fe2	1173	1223	65	5	20	10	12.5	5.1	10
Fe3	1173	1223	60	5	20	15	12.5	3.6	15
Fe4	1173	1223	55	5	20	20	12.5	2.7	18

^a f_s = solids circulation rate; ^b ϕ = oxygen carrier-to-fuel ratio