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Distribution and treatment of harmful gas from heavy oil production in the Liaohe Oilfield, Northeast China

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Abstract: The distribution and treatment of harmful gas (H_2S) in the Liaohe Oilfield, Northeast China, were investigated in this study. It was found that abundant toxic gas (H_2S) is generated in thermal recovery of heavy oil. The H_2S gas is mainly formed during thermochemical sulfate reduction (TSR) occurring in oil reservoirs or the thermal decomposition of sulfocompounds (TDS) in crude oil. H_2S generation is controlled by thermal recovery time, temperature and the injected chemical compounds. The quantity of SO_4^{2-} in the injected compounds is the most influencing factor for the rate of TSR reaction. Therefore, for prevention of H_2S formation, periodic and effective monitoring should be undertaken and adequate H_2S absorbent should also be provided during thermal recovery of heavy oil. The result suggests that great efforts should be made to reduce the SO_4^{2-} source in heavy oil recovery, so as to restrain H_2S generation in reservoirs. In situ burning or desulfurizer adsorption are suggested to reduce H_2S levels. Prediction and prevention of H_2S are important in heavy oil production. This will minimize environmental and human health risks, as well as equipment corrosion.

Key words: Toxic gas, H_2S, heavy oil production, TSR, Liaohe Oilfield

1 Introduction

As a highly toxic and corrosive gas, H_2S is an important safety concern in production because it can cause great damage to the surroundings and human beings (Orr, 1977; Machel et al, 1995; Krouse et al, 1988; Worden et al, 1995; Dai et al, 2004; Zhu et al, 2005a). A concentration of H_2S lower than 0.2-0.3 ppm is safe for human beings. There will be a strong smell when at 20-30 ppm. Olfactory paralysis will occur when its concentration is 100-150 ppm. People can die within a couple of seconds when the H_2S concentration is 1000 ppm. The H_2S concentration must be less than 1 ppm according to the national standard of China (GBZ 2-2002). In most blocks in the heavy oil steam drive areas of the Liaohe oilfield, the concentration of H_2S in natural gas is more than 200 ppm. This level is dangerous for people.

During the oil exploration and development processes in China, there have been more than 10 major accidents from H_2S toxicity, resulting in great casualties and property loss. Therefore, the concentration of H_2S gas has been monitored since these accidents happened. It has been reported that large amounts of H_2S is generated during heavy oil thermal recovery process (Aplin et al, 1995; Wilhelm, 1981; Lamoureux-Var and Lorant, 2005). It is a new problem, first found in the Liaohe Oilfield, in the thermal recovery process in China.

The Liaohe Oilfield is the third largest oilfield in China, and also the largest production base of heavy oil and extra-heavy oil in China. Development of the heavy oil resource and its enhanced oil recovery (EOR) is the focus for the recent exploration and development of the Liaohe Oilfield. As a main method in the early development stages of Liaohe oilfield, steam soak has a history for more than 20 years, and the rate of crude oil production declines with the steam soak times. As a follow-up method, steam drive was first applied in Gaosheng area in 1998, and then was applied in the Qi 40, Wa 38 and other pilot areas in the Liaohe Oilfield. In recent years, toxic gas, mainly H_2S, has been found in many pilot areas, especially, with a higher concentration in the steam drive areas. For example, the concentration is up to 4.49% in well Wa-3730 of Wa-38 block in the Xiaowa area, and up to 3.27% in well Qi 40-9-025 in Qi 40 block. And gas with low

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H_2S content has also been found in other thermal recovery pilot areas, oil and gas gathering equipment and transportation pipelines, and multi-function stations.

2 Generation of H_2S in thermal recovery pilot areas of heavy oil in the Liaohe Oilfield

H_2S was first found in gas in the late 1990s during the steam drive process in Gao 3 block of Gaosheng Oilfield in the western depression of the Liaohe Oilfield. The content of H_2S is between 0.41% and 0.95%. In recent years, H_2S has been found in the Qi 40 and Wa 38 blocks (seen in Fig. 1). Besides, low content H_2S has also been found in some steam soak areas. Judging from the current H_2S distribution, it is mainly located in the heavy oil areas in the western depression of the Liaohe Oilfield, particularly in the steam drive areas.

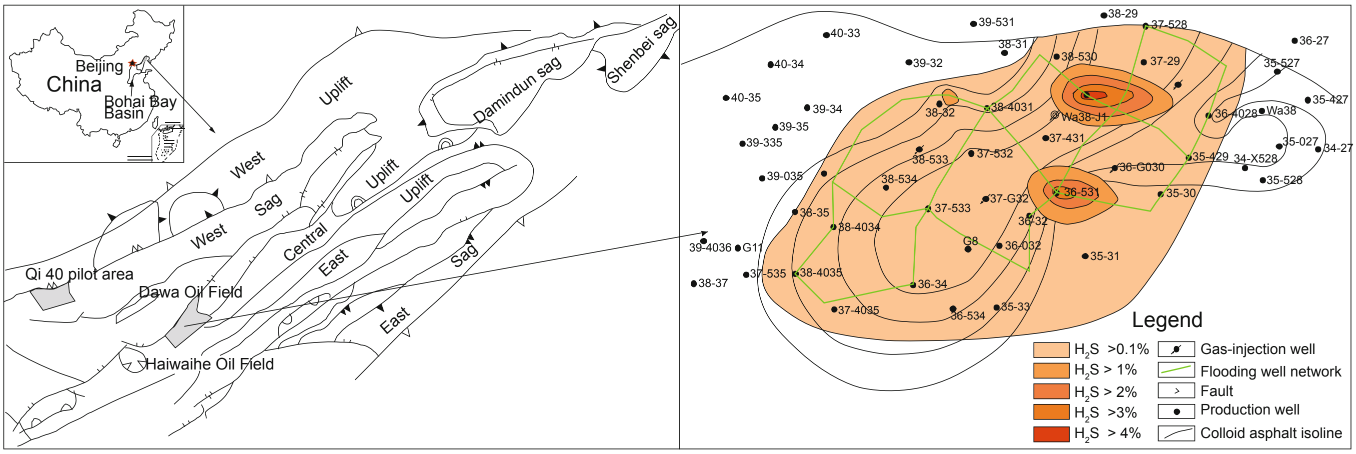

Fig. 1 Distribution relationship between hydrogen sulfide and colloid & asphaltum in the Wa 38 block, Liaohe Oilfield

Judging from the H_2S distribution, nearly all the H_2S was formed during the thermal recovery process, so it is of secondary origin. Therefore, we could use the exclusive method to make a preliminary determination. H_2S formed from bacterial sulfate reduction (BSR) has a wide range of distribution (Machel, 2001), however, this kind of H_2S-containing gas has some distinctive characteristics: First, H_2S content is low; Second, the reservoir conditions have to be suitable for micro-organisms to live, especially the temperature. It is known that the maximum temperature for most micro-organisms is about 80 ºC, that is, if the temperature is higher than 80 ºC, all the micro-organisms, especially the sulfate-reducing bacteria, will die. Under normal conditions, the most appropriate temperature for sulfate-reducing bacteria to live is about 34 ºC. The reservoir temperature in thermal recovery pilot areas of Liaohe heavy oil is above 80 ºC, which is higher than tolerated by sulfate-reducing bacteria, so we can exclude biogenic H_2S as a source for the H_2S in this area.

Sulfur-bearing organic material occurs in oil and gas reservoirs, for example, in source rocks and crude oil. The organic matter mainly exists in crude oil in Liaohe heavy oil province, and the sulfur content is between 0.1% and 0.4% (low-sulfur crude oil). When sulfur compounds are heated, heterocyclic sulfur cracks and H_2S is formed, that is, thermal decomposition of organic matter. However, H_2S formed in this way could not reach high content. The low H_2S content is likely to be related to thermal decomposition of the sulfur-bearing organic compunds.

For H_2S formation, gypsum or other sulfate minerals, hydrocarbons and high-temperature conditions are required for TSR (Zhu et al, 2005b; 2007; Mougin et al, 2007). The thermal recovery temperature in the heavy oil reservoirs of the Liaohe Oilfield is higher than 120 ºC, suitable for TSR. However, no significant gypsum is found in the Shahejie formation, so many scholars have questioned the causes of TSR (Zhang et al, 2005; Zhu et al, 2005b). In fact, when TSR occurs, it is the thermal chemical reaction between crude oil and SO_4^{2-} rather than the reaction between crude oil and gypsum, that is to say, a certain concentration of SO_4^{2-} in the formation water is required. The investigation in this paper shows that the high SO_4^{2-} content, usually hundreds of parts of per million, in the formation water of the Liaohe oilfield is the basis for TSR to produce H_2S.

Previous research (Zhu et al, 2009) shows that H_2S is mainly generated from the TSR (thermochemical sulfate reduction, i.e. hydrocarbon and sulfate ions react chemically) and from the thermal decomposition of sulfocompounds (TDS) within oil in heavy oil pilot areas of the Liaohe Oilfield. Although hydrothermal cracking of heavy oil can produce H_2S with a wide range of distribution during the stream drive process, the H_2S content is usually low. Sulfate required for TSR comes partly from the chemical reagent injected into the reservoirs before and after the thermal recovery and partly from high levels in the formation water in the third section of Shahejie formation. The thermal recovery temperature is also suitable for TSR, so TSR is probably taking place in the reservoir. Besides, because of the poor thermal stability of some injected chemical reagents (such as sulfonate), H_2S can be formed by their thermal decomposition during the steam drive process.

3 H_2S distribution in the thermal recovery pilot areas of heavy oil in Liaohe Oilfield

The data analysis on the H_2S-containing gas made by the Institute of Oilfield Test Center and the Huanxiling oil production plant, Liaohe Oilfield, shows that H_2S distribution is wider in heavy oil pilot areas than in light oil pilot areas. In the heavy oil pilot areas, wells with high H_2S content are all in the steam drive areas (Fig. 2). In the Qi 40 block, there are 110 wells with H_2S content above 200 mg/m^3, accounting for about one-third of the total wells. Though the total horizontal H_2S distribution is not very clear, the wells in the central Qi 40 steam drive area all have a high H_2S content, indicating that high-temperature steam drive may be favorable to H_2S generation, and the high-temperature is likely to be an important factor for high H_2S concentration.

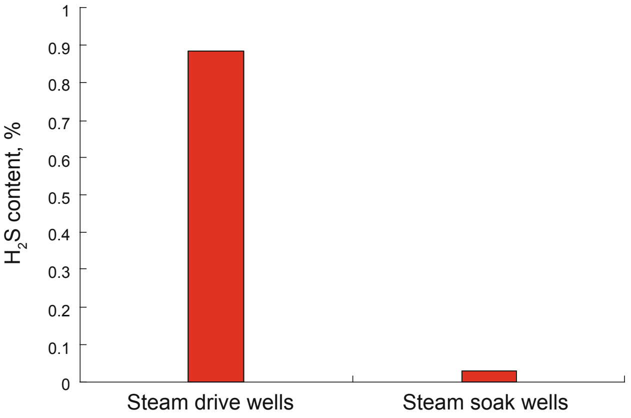

Fig. 2 Comparison of H_2S contents between steam drive wells and steam soak wells

4 Relationship between H_2S content and recovery time and mode

Based on the H_2S distribution in each area (seen in Fig. 1), the distribution of H_2S is wider in the stream soak pilot area than in the stream drive area. However, the H_2S content is higher in the stream drive area than in the steam soak area, indicating that H_2S generation is related to the mode of thermal recovery.

In the thermal recovery pilot areas of heavy oil in the Liaohe Oilfield, the area of high H_2S content often locates in the pilot test area in comparison to the expanded pilot area. Two areas of the Qi 40 block are now in the steam drive stage, and different steam drive durations have led to different H_2S concentrations. In general, the H_2S content is higher in the pilot test area than in the expanded pilot area. The high H_2S content also appears in the pilot test area and its peripheral areas.

Table 1 shows the test result of H_2S for well Qi 40-9-028, monitored by the Institute of Oilfield Test Center, Liaohe Oilfield, in the expanded test area. It is seen that the H_2S content was up to 0.79% and fluctuated during the development process, which might be related to the EOR operations.

Table 1 Changes in H_2S content for Qi 40-9-028 well in steam drive pilot area

Monitoring frequency	H_2S v/v, %	H_2S mg/m^3	Monitoring date
1	0.2779	3986	2005-11-05
2	0.7389	10599	2006-03-23
3	0.7448	10683	2006-03-24
4	0.6306	9045	2006-06-06
5	0.7893	11321	2006-08-21
6	0.2352	3374	2006-11-02
7	1.2162	17445	2006-12-05

The results of the seven tests are not stable, with an upward trend. In November 2005, the measured H_2S concentration was 0.2779%, and after profile control, on December 5, 2006 it was up to 1.2162%. As the profile control increased the spread of injected steam, some operations in the production process, such as steam-soak after steam injection, profile control and viscosity reduction, would be favorable to increase the concentration of H_2S. Based on the monitoring of the gas in the wells, it is found that the concentration of H_2S increased in the expanded pilot area along with the time of extended steam drive. In addition, the concentration of H_2S kept an upward trend as the exploitation time is relatively short in extended test area. There is no obvious pattern in the change of H_2S levels with time in the pilot test area. However, the time of steam drive controls the production of H_2S to some extent.

5 Main influencing factors on H_2S concentration

The concentration of H_2S in well gases varies significantly in the same area or under the similar reservoir conditions. Besides the geological conditions or geological backgrounds, the TSR reaction is probably the most important factor for the difference of H_2S concentration. Generally speaking, the TSR reaction is more likely to happen with an increase of depth and temperature. In addition, the amount of SO_4^{2-} in the formation water is also an important influencing factor for the TSR reaction. If the SO_4^{2-} levels drop substantially, the TSR reaction will stop.

Fig. 3 shows that the formation water collected in Qi 40 block is rich in SO_4^{2-}, which offers a source of reagent for TSR reaction and the concentration of SO_4^{2-} mostly reduced after the thermal reaction. The reason is probably that TSR reaction has, in many cases, consumed most of the SO_4^{2-}.

In addition, both the heating level of the reservoir and the duration of its high temperature affect the generation of H_2S gas. Usually, the content of H_2S gas becomes higher and higher with increasing reservoir heating time, indicating that the TSR reaction plays an important role in the generation of H_2S.

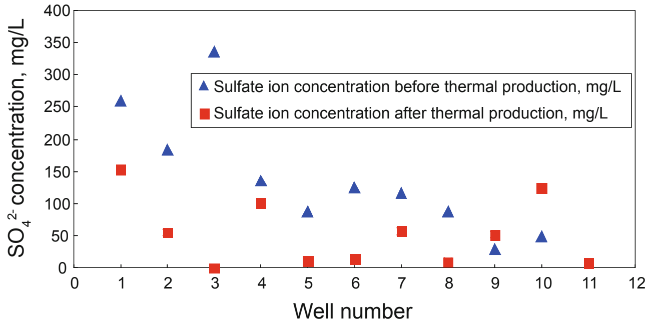

Fig. 3 Change of SO_4^{2-} in the formation water before and after thermal production for the Qi 40 block

6 Distribution of H_2S

Considering the H_2S formation conditions and its control factors, H_2S will be generated in most oil wells in heavy oil thermal recovery areas and become a serious problem for safety and environmental protection if there are no countermeasures taken. In the thermal recovery region and in the absence of naturally occurring SO_4^{2-} in the formation water, if no chemical reagents with SO_4^{2-} were injected into the formation water, the TSR reaction would not take place. Hence, there will be little H_2S generated. H_2S from the thermal decomposition of crude oil is quite low, and will stop generating when the thermal decomposition of sulfocompound (TDS) in crude oil is finished. But the content of H_2S gas will increase if there is no measure to take in both the pilot areas and the expanded pilot areas. Taking the Qi 40 block for example, the pilot area with high H_2S content have low SO_4^{2-} content because SO_4^{2-} has been consumed by the TSR reaction, but the SO_4^{2-} content is still high in the peripherals of the pilot area, the H_2S content in the peripheral area will probably increase in the near future.

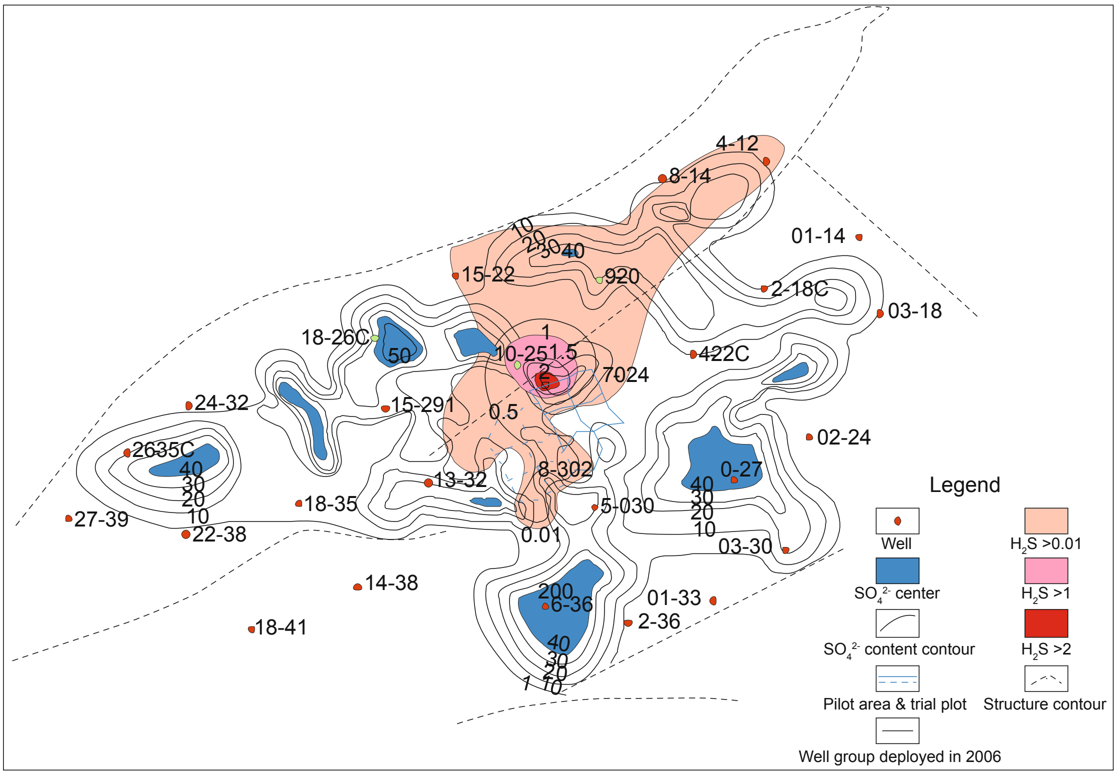

Fig. 4 Relationship between SO_4^{2-} and H_2S distribution in the Qi 40 block

7 Equipment damage from H_2S

Due to its strong chemical activity, H_2S is extremely corrosive to drill tools, gathering and transportation pipelines and so on. In addition, it can also cause “hydrogen brittleness”. The corrosion will increase when H_2O, CO_2 and O_2 exist in the environment. Even low concentrations of H_2S can react with Fe to form a compact FeS film, which is mainly composed of FeS and FeS_2. With increasing concentration of H_2S, the FeS film becomes black loose layers or powders, that is, $Fe+H_2S \rightarrow H_2+FeS$. Hydrogen brittleness and sulfide stress corrosion cracking are important mechanisms of metal corrosion, which is the one of the main reasons leading to major accidents. Corrosion perforation probably happens in plenty of casing and consequently underground pipelines would be corroded. Sucker, oil pumps and many pipelines are scrapped due to corrosion each year. The maintenance and replacement of pipelines cost tens of millions of dollars because of corrosion. The content of H_2S is usually more than 500 mg/L in this equipment. Major accidents will happen, such as pipeline bursts, if no measures are taken to control the high levels of H_2S.

8 Harm prevention measures from H_2S in thermal recovery pilot areas of heavy oil in the Liaohe Oilfield

8.1 Periodic examination

The components, structure of gaseous pollutants, especially H_2S must be monitored in the thermal recovery pilot areas of heavy oil. A unified, well-founded monitoring system for the main facilities should be developed in the heavy oil thermal recovery. The system should be able to monitor and identify if there is H_2S in the wellhead, oil, water tanks, purification tank, and other facilities or not. In addition, the monitoring system could make clear the quantitative relationship between H_2S emissions in the thermal recovery of heavy oil and the technologic process and meteorological parameters. All these measures are taken for preventing serious personnel injury and equipment damage due to high content of H_2S.

8.2 Making people aware of the harm of H_2S gas

Much more should be done to make people to realize the risk and harm of H_2S. Meanwhile, the warning signs and danger signs of H_2S should be pasted in the places where H_2S is usually generated at dangerous levels.

8.3 Removing H_2S by absorption

Plenty of liquids can be used as absorbents for H_2S, such as neovaricaine, tripotassium phosphate, and sodium salt. Neovaricaine is the most efficient absorbent because of its high absorption rate, low freezing temperature and flexible equipment requirements. Each aqueous solution of monoethanolamine, diethanolamine and triethanolamine can also be used to remove H_2S.

8.4 Proper measures for protecting pipeline materials

The use of nickel-phosphorous plating technology in pipeline materials is one preferred measure to protect the pipelines in the heavy oil thermal recovery. It can prevent or delay pipeline corrosion during the heavy oil thermal recovery process. The corrosion rate could be reduced by 50% to 70%, which means that the working life of pipelines could be extended up to two or three times. In addition, pulse vacuum nitriding coating technology of tubes to resist corrosion is a useful chemical surface treatment technology. After the chemical surface treatment, the surface corrosion resistance of the pipelines could be greatly reduced as a dense white layer of corrosion resistance formed on the entire surface, without reducing the original mechanical properties.

9 Treatment measures for H_2S in thermal recovery pilot areas of heavy oil in the Liaohe Oilfield

9.1 The present disposal method

In the Huanxiling oil production plant of the Qi 40 pilot areas, measures for H_2S disposal are mainly venting at high altitude and control of casing pressure. Control of casing pressure can reduce emission of H_2S, but it has a great influence on oil production. Oil pumps may suffer from gas locking when the casing gas gets into the pump, so it will affect the oil production. The Liaohe Oilfields have conducted casing control tests for the Qi 40-9-027 and 40-9-028 wells, and found that the daily oil production of Qi 40-9-028 well dropped from 12 m^3 to 4 m^3 after casing pressure control. The daily oil production of the Qi 40-9-027 well is still 4 m^3, so the influence is small for Qi 40-9-027 well. H_2S was not removed after casing control, but ran into the system. This would result in the high concentration of H_2S in the transportation system.

At present, venting at high altitude is a good way for disposal H_2S, but once the Qi 40 block comes into the stream drive stage, a large-amount of H_2S would be formed. Although the air can dilute the H_2S, H_2S concentration on the ground will still exceed the limit as the density of H_2S is higher than air. Therefore, this method would not solve the problem.

9.2 Restraining the generation of H_2S

It is known that H_2S is generated mainly from the thermal decomposition of sulfocompounds (TDS) within oil and from the TSR reaction. H_2S formed by TDS is much less, and sulfur compounds are used up after a long period of thermal recovery. So TDS plays less dominant role after a period of thermal recovery. TSR is mainly controlled by temperature and SO_4^{2-} content. The temperature cannot be changed, so we could take measures to reduce SO_4^{2-} content, for example, by not injecting SO_4^{2-} bearing chemical reagents, thus, TSR reaction will be stopped or diminished due to lack of reactants, and the generation of H_2S will be stopped. Therefore, the key step for restraining the generation of H_2S is to control injection of SO_4^{2-} bearing compounds.

9.3 Not using sulfur compounds with thermal instability

Before and after the steam drive, foaming agents, demulsifiers and other materials with sulfate ions or sulfonic acid ions have been injected into the reservoirs, some of which may be thermally unstable. So thermal decomposition can occur during the thermal recovery process and produce H_2S. In spite of the distribution of H_2S being limited, it is very difficult to predict the distribution area of H_2S. Also, it is difficult to predict the change of content of H_2S. Therefore, the thermal stability performance of the reagents should be examined first before they are injected into the reservoir.

9.4 In-situ burning of H_2S

H_2S is a combustible gas. In-situ burning of H_2S will reduce its harm. However, in the casing tube of Qi 40 block, the content of hydrocarbons is low, while the content of carbon dioxide is about 80%-95%. Only a few wells have a low content (about 20%) of carbon dioxide in the casing tube. Therefore, most of the oil well casing gas cannot be combusted directly. It needs to add natural gas for burning H_2S. The reaction is: $2H_2S+3O_2\rightarrow 2H_2O+2SO_2$. It would be better to use a chimney in this method as it can cause gas pressure different, which is helpful for burning. In addition, the combustion products can be diffused high in the air, and the harm is small. This method has the advantage of simple construction, small and flexible transformation process and low investment. It can be used not only for the single well but also for a platform or the oil wells nearby. For the Qi 40 block, the processes in the whole block can be completely transformed in 2 to 3 weeks. The limitation of this method is that SO_2 generated in the process is harmful to the environment. In addition, the contents of hydrocarbons and carbon dioxide often fluctuate, so the natural gas to casing gas ratio is not easy to control, that is, if not enough natural gas is added, the combustion of the mixed gas will be incomplete. While if too much natural gas is added, energy and gas is wasted. However, this method can be used for combustible gas, to avoid loss of gas in the casing tube. For the gas that cannot be ignited, desulfurating agent adsorption technology can be used instead.

9.5 Centralized disposal in a desulfurization plant

In the heavy oil areas, the generation of H_2S cannot be controlled when using plenty of steam. A small-scale desulfurization plant for disposal of H_2S should be established in a suitable location. The desulfurization technology is relatively mature in China, for either dry or wet desulfurization technology. In addition, there is a lot of experience in the manufacture, use, and maintenance of such equipment in China.

10 Conclusions

Large amounts of toxic H_2S gas is generated during the process of thermal recovery of heavy oil in the Liaohe Oilfield, China. The gas is mainly formed by the thermochemical sulfate reduction (TSR) occurring in the oil reservoirs. Also, it can be formed by the thermal decomposition of sulfocompound (TDS) in crude oil. The generation and distribution of H_2S gas are controlled by the time and style of exploitation, temperature in the thermal recovery and the injected chemical reagents.

SO_4^{2-} is the necessary material for TSR reaction. The supply rate of SO_4^{2-} (SO_4^{2-} content in the formation water) is the most influencing factor for the rate of TSR reaction. The content of SO_4^{2-} of the boiled water injected into the reservoir is high, which provided SO_4^{2-} source for the TSR reaction. It is also the reason that content of H_2S is relatively high in the pilot areas.

In the process of H_2S prevention, periodic and effective monitoring should be undertaken in the heavy oil thermal recovery pilot areas. In addition, H_2S gas absorbents and corrosion resistant pipeline material should also be used. We should make every effort to control the generation of H_2S in the heavy oil recovery areas in the beginning, such as cutting off the supply of sulfate to the TSR reaction. The method of burning in the well field or adsorption by desulfurizer could also be considered for the disposal of H_2S gas.

In conclusion, the distribution area and concentration of H_2S gradually increases along with extension of heavy oil thermal recovery area. As a result, the potential safety hazard resulted from H_2S becomes larger and larger. It will result in higher risk for exploitation of heavy oil. Also, it becomes challenging for the health of workers and protection of equipment. Therefore, it will produce good social benefits such as stable production, better occupational health levels, protection of equipment from corrosion and environmental protection if the dynamic distribution in situ of H_2S is well researched and measures to prevent the formation of H_2S are taken.

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