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POTENTIALS OF SILICATES IN TREATMENT OF OIL PRODUCING WELLS

By joint application of silicates and polymers, a multifunctional, self-controlling chemical system is formed which works spontaneously even under harsh reservoir conditions, meanwhile the methods remain inexpensive, flexible and adaptable to any production technologies. A concise summary of the technique, its principle and mainly field projects are discussed in the paper. It was shown that the silicates, combined with polymers offer unique opportunity to cure numerous production/injection problems including water-shut-off, profile correction, gas coning, etc. in oil and gas fields. Between 1980 and 1998 the field projects, comprising more than forty well treatments, yielded substantial additional oil production, life time of wells were extended and the overall profitability of the field was significantly increased, meanwhile environmentally friendly chemicals were used. Therefore, the Hungarian experts are convinced that the polymer/silicate method is reasonable alternative wherever and whenever the application of bulk or surface gelation or deposition is arising.

INTRODUCTION

In hydrocarbon industry the silicates as diverting and sealing agents were first proposed as early as the twenties. The advantageous features of silicates were reinvented only at the end of the sixties, although their field applications were and still is shaded by the different polymer gels. Thus, despite some remarkable positive field pilots, the use of silicates in water shut-off treatments, profile corrections, clay stabilization, etc. remained marginal until now.

Hungary ranks among the countries where the silicates not only occasionally, but continuously are applied under field conditions. During the past two decades the silicate-based methods were used for water shut-of in oil producing wells, profile correction in water injection wells, restriction of gas coning, stabilization of reservoir rocks, inhibition of clay disintegration, etc. More than a hundred wells were treated and as a result some hundred thousand tons of surplus oil production was registered, productivity and injectivity of wells were temporarily or permanently improved, after all the profitability of oil production was significantly increased. The paper gives a concise summary of the polymer/silicate technique, outlines its principle and demonstrates some characteristic field trial. It will also be shown that the silicates, as main component of gels and treating solutions, offer unique opportunity to cure numerous problems solved conventionally by organic gel-forming materials.

THEORETICAL APPROACH

The idea of water shut-off treatments or local profile correction in vicinity of wells raised as early as the middle of the sixties. Since that time a great variety of so-called polymer methods have been developed forming three main groups: *injection of polymer solution*, *in-situ cross-linking of chain like polymers* and *in-situ polymerization of monomers*. Their general feature is that they are based on application of macromolecular materials. Such a common denominator can not be found as far the blocking mechanism is concerned. Since the philosophy of profile correction methods is that a deliberate formation damage or a drastic mobility reducing effect must be developed in a right reservoir space and in a proper stage of production, the techniques, in a wider sense, can be classified as follows:

Mobility control by modification of rheological properties

- Injection of polymer solutions;
- In-situ cross-linking of linear polymer;
- In-situ polymerization of monomers;
- Combined, multifunctional methods;
- Precipitation of gel-like inorganic compounds;
- Precipitation of crystalline inorganic compounds.

Mobility control by modification of pore structure

Theoretically, the reservoir engineering concept of the profile correction or in-situ barrier formation is invariant to the factors whether the viscosity of fluids, pore structure the reservoir or simultaneously both of them are modified in the target area. Positive and negative arguments may aptly be listed beside all solutions. Surprisingly, however, the profile correction techniques based on joint application of inorganic compounds (e.g. silicates, hydroxides, humates, etc.) and polymers, particularly those which aimed at permeability lowering through reducing the free cross-section of flow, were practically ignored for a long time. Therefore, the primary aim of the research project comprising more than two decades in Hungary, was to analyze these alternatives and to evaluate the potential of such methods under field conditions. The main guideline of the efforts was that the flexibility, reliability and efficiency of the new procedures must be superior to conventional techniques.

BACKGROUND OF SILICATE-BASED METHODS

Application of silicates in different industrial areas is wide-spread and enormous number of papers, books, etc. deal with the theoretical and practical aspects of behavior, structure and formation of silicate gels. Injection of silicate solutions into reservoirs with the aim at enhancing the recovery factor through a diverting effect was proposed first by Hill [1] claiming a patent in 1922. Looking back to the many decades of research and industrial utilization we may conclude that despite intensive patent activity between 1940 and 1980 the practical application of silicates in frame of IOR/EOR technologies is rather an exception than a generality. The primary target area of silicate technology in the mentioned period, as detailed by Baker [2], was the civil engineering, where the silicates were extensively used as grouting agents. Robertson and Oefelein [3] were one of the few who not only studied, but also tested the silicate gels for water shut-off. Later, and Cole et al. [4, 5] and Sparlin at al. [6] presented some papers proving that the traditional grouting materials might be used successfully for IOR/EOR purposes. At this time the extension of applications was already supported by excellent comprehensive papers and books [7-10], which were treasury of updated information on silicates.

In literature, the paper published by Krumrine and Boyce [11] might be considered as a milestone, because they gave not only an almost complete summary of the topic listing numerous papers and patents, but they also drew attention to a controversial fact that the silicates were inequitably neglected beside polymers in practice. The authors expressed their firm conviction that the "permeability modification with silicate gel-based system is a viable alternative wherever the need arises". That statement is rooted in arguments that the different silicate systems are flexible, the water soluble silicates can be gelled by a great variety of inorganic and organic compounds, and natural materials, and thus, the methods might be tailored and adapted to diverse reservoir conditions.

The reasons, why the silicates have not been used more widely in practice, were hardly understood for a long time, but at the same time, the intriguing questions stimulated further lab studies. Vinot et al. [12] listed four possible reasons, and among others, they pointed out that the mechanism of silicate gelation, particularly under reservoir conditions, is poorly understood. Taking their conclusions also into account, the pros and cons of silicate well treatment techniques can be summarized as follows:

Advantages:

- Low viscosity of treating solutions, viz. good placement selectivity;
- Short to moderate pumping time before onset of gelation;
- Flexible chemical mechanism;
- Good chemical and chemical stability;
- Excellent thermal and mechanical resistivity;
- Easy gel breaking in case of technical failures;
- Simple and cost-effective surface technology; and
- The silicates are environmentally friendly materials.

Disadvantages:

- The gel is rigid and prone to fracture;
- The gel shows syneresis, viz. it is prone to shrink;
- Because of shrinking the blocking efficiency is changing in time, and it is never total;
- Penetration of the treating solutions is short if the buffer capacity of rock is high;
- The setting time is usually short, namely, it is hard to control the gelation mechanism;
- Silicates are prone to form precipitates instead of gel;
- Alkaline silicates initiate intensive ion exchange; and hence, precipitation of multivalent cations; and
- Silicates change the interfacial properties, thus they enhance in-situ colloid chemical processes (emulsification, agglomeration, etc.)

Recognizing the factors influencing unfavorably the properties of silicate gels, different solutions were reported in literature to overcome the mentioned shortcomings. Mixture of alkaline silicate and acid phosphate solution was proposed by Beecroft and Maier [13], while a more sophisticated techniques, a complex solution containing polymers, cross-linkers, silicates and gelling agents were patented by Sandiford [14]. Vinot et al. [12] have shown that applying hydrolizable esters, introduced into the alkaline silicate solution as a dispersed phase (microemulsion), the gelation of system ensued even though the pH remained constant (>11). They also stated that as a result of a unique gelation mechanism, the properties of gel and the chance to have permanent and efficient barrier formation under harsh reservoir conditions were significantly improved. These are only representative samples of efforts and today it is hard to overview completely all theoretical and practical varieties of methods, which are basically *silicate* techniques.

In Hungary, the silicate type well treatment methods were intensively studied and tested at oil fields from the middle of the seventies. In principle, three different versions were developed:

1. Addition of anionic polymers to silicates with the aim at simultaneous cross-linking of polymers and gelation of silicates resulting in a double network consisting of a rigid and a flexible chain in the gel [15-17].
2. Cross-linking of silicates by cationic polymers in a bulk phase resulting in a semi-flexible, single network in gel [18].
3. Gelation of silicates in the presence of additives, mostly humates, resulting a rigid, but clustered network in gel [19].

These technical solutions are addressing not all, but some of the unfavorable properties of gel, and the main and subgroups of the developed procedures may offer, and as proved by pilot tests, substantial improvements if the chemical systems are appropriately formulated. The disproportional permeability reduction (DPR) by silicates is, however, not limited to Hungary. As Schilling [20, 21] recently reported the pure silicate method was successfully applied at the North See and as a result of two treatments 130,000 t of additional oil production is predicted at the Gullfaks field until 2000. In an other paper the same authors [22] stated however that DRP, independently of the gel forming materials, may not cure all problems and sometimes the results are negligible. After all, despite some contradicting field results, we may accept as a directing opinion disseminated by Pusch [23], that silicates provide a reasonable alternative for profile correction in harsh reservoir environments (>150 °C) when other chemicals, particularly polymer gels, may not be used.

PRINCIPLE OF THE POLYMER/SILICATE METHODS

Development of a well treatment technique based on joint applications of polyacrylamides and silicates goes back to the end of the seventies. At this time the primary initiatives were to elaborate a method which can be used for restriction of water influx to bottom holes in oil producing wells. Later on the overall profile correction in vertically heterogeneous reservoirs was the basic task to be solved. Parallel with this efforts, in a special technical frame, the polymer/silicate barrier forming system became also a relevant part of a so called foam/gel complex method tested several times in field for restriction of gas coning [24, 25]. In the meantime, a modified version of the technique was successfully applied for complete blocking of vertical gas migration in damaged wells [18]. The laboratory and field studies were also accelerated by new incentives on application of the same system as a sealing agent in environmental technology [26] and fundamental research connected with the transport phenomena of cross-linking multivalent ions, hydrogen ions and other species in polymer/silicate gels [27-29].

The technique is a multifunctional profile correction method based on simultaneous cross-linking and polymerization of hydrolyzed polyacrylamide and sodium orthosilicate, respectively. This procedure was already detailed in earlier publications [15, 16]. The high efficiency of its chemical mechanism is attributed to parallel chemical reactions taking place in the mixing zone of treating solutions which contain the following main components:

Solution A : partially hydrolyzed polyacrylamide (PHPAA)
silicate (Na or K ortho-silicate)
Solution B : potassium aluminum sulfate (alum)
calcium chloride
hydrochloric acid

Generally, the composition of treating fluids are as follows:

Solution A : 1-2 g/l PHPAA
10-50 g/l SiO_2 containing water glass
Solution B : 2-5 g/l alum
2-5 g/l $CaCl_2$

The concentration of hydrochloric acid in Solution B may vary between wide limits and depends on many factors (penetration depth, buffer capacity and acid soluble content of rocks, concentration and type of commercial silicate, etc. In addition, the solution may contain different stabilizers, inhibitors, etc. and the main components can be replaced by numerous other chemicals corresponding functionally to the basic tasks. These solutions might be used in form of a sequential or a bulk phase injection scheme depending on the desired penetration depth. Since the gelation time can be controlled within certain limits only, sequential injection is usually preferred over bull head and bulk phase injection.

In the mixing zone or phase of the mentioned solutions, particularly if it happens in natural porous media, the following processes are taking place:

Chemical reactions : cross-linking of polymers
gelation (polymerization) of silicates
precipitation of
polymers
silicates and
aluminum hydroxides
Interfacial effects : adsorption of chemicals
entrapment of microdomains
(microgels, clusters, hydroxides, etc).

The parallel and coinciding processes jointly result in a radical mobility reduction in the target reservoir space, but contributions of rheological and the pore structure modifying factors (actual permeability reduction) significantly depend on the local conditions, the chemical system and the placement technique applied. Under favorable or optimal circumstances the blocking efficiency of the polymer/silicate gels may be characterized by the following parameters:

Permeability reduction : 4-5 orders of magnitude
(from 1 D to 0.1-1.0 mD)
Gel strength : >10 bar/m
Thermal stability : >150 °C
Mass transport in gel : diffusion
Blocking barrier : sandwich type (if sequential injection is used).

Composition and properties of the gels itself and the complex chemical reactions, leading to permeability reduction, imply that most of the unfavorable features of a pure silicate gel can be omitted by joint application of polymers and silicates. Formation of a double network in the gel is in the background of improvements, but existence of both the controlled and the spontaneous elements in the chemical mechanism may provide further attractive features, like

- flexibility to formulate the treating solutions, to design the surface technology and to tailor the method to great variety of field conditions;
- excellent chemical, thermal, mechanical and microbial stability with reduced propensity to shrink (syneresis);
- high gel strength (elasticity) which is superior to both the pure polymer and the silicate gels;
- simple surface technology, eliminating all difficulties connected with manipulation of high molecular weight polymers (dissolution, filtering, shearless facilities, special surface coatings, etc.)
- cost and labor effective procedure easy to control and supervise; and
- less environmental impact on well site.

These positive features were properly proved and demonstrated by the pilot treatments and routine applications at the fields.

FIELD STUDIES AND ROUTINE APPLICATIONS

The field tests and routine application of methods based on silicates were performed at the largest Hungarian oil and gas field. The Algyő field, discovered in the early sixties in south Hungary, is the largest Hungarian hydrocarbon field. Special feature of the multilayered reservoir consisting of more than 70 independent hydrodynamic units, is that some layers have measurable gas cap. Table 1. lists the basic parameters of the system. Production at the field began in 1965. In the primary stage only the formation energy was used. Later, the production technique included partial water flooding and gas lifting. Today, double-sided water flooding and extensive gas lifting are the mainstays of the technology. In the primary stage 5-10 % recovery factor was attained, and it is thought that the recovery efficiency today exceeds 40 % OOIP. Between 1970 and 1995 the annual oil production has been permanently over 1 Mm^3/y (Fig. 1.). Since 1990, however, the annual oil production shows decreasing tendency. In 1995 the oil production totaled only at about 0.7 Mt/y. The water production started at the field in 1971 and it reached top rate with 4-5 Mm^3/y between 1989 and 1994. It is characteristic that the annual oil and water production became equal at the end of the seventies and after that the water production doubled and tripled. On account of time lag and the steeper trend of water production the cumulative curves intersect each other indicating that two times more water was produced until now than oil (Fig. 1).

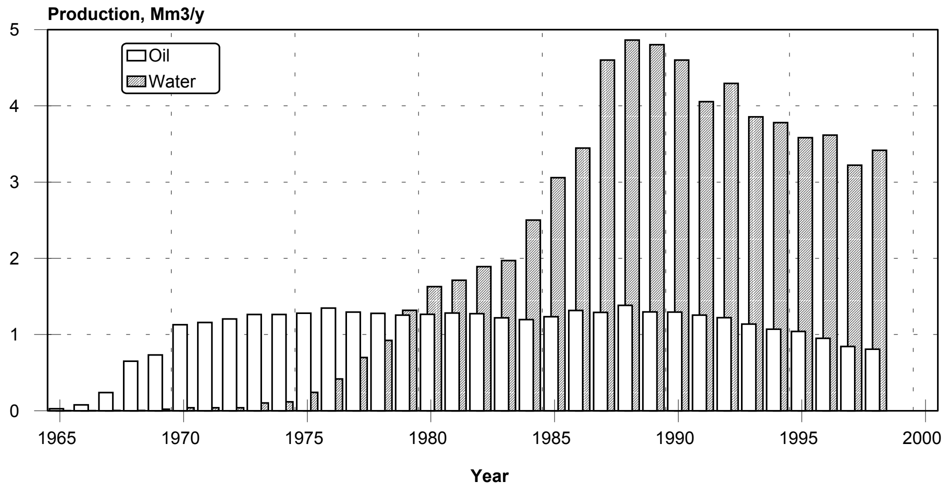

Fig. 1. The annual oil and water production at the Algyő field, Hungary

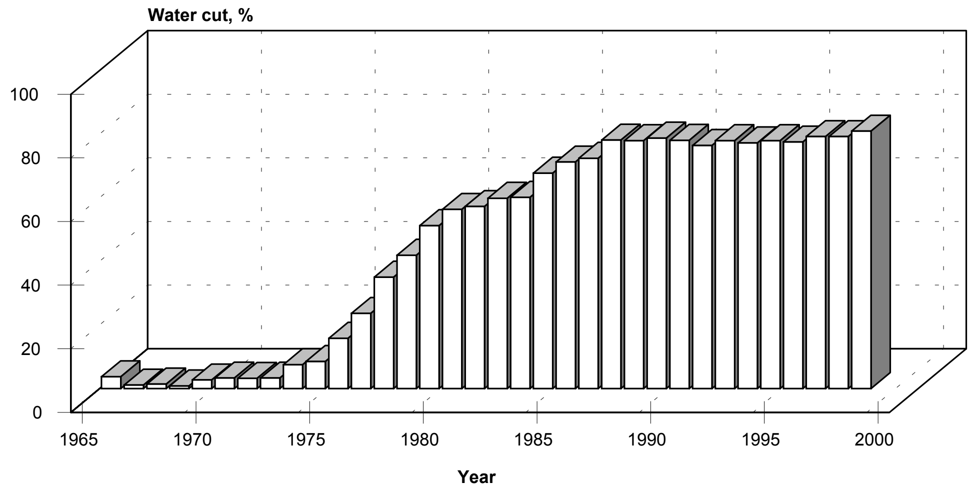

Fig. 2. Change of the average water cut of oil production at the Algyő field, Hungary

The water injection at the field started in 1975 and reached its maximum in 1989 with 8-9 Mm^3/y. Consequently, the oil production was practically free of water until 1973, then the water production gradually increased in the period of 1975-1988 (Fig. 1). At the present time the average water cut stabilized at roughly 80 %, but on account of the declining oil production it shows slightly rising tendency (Fig. 2). Unfortunately, there are hundreds of wells which already operate with water cut higher than 90 %. Therefore, one of the basic problems, the operator has to face with, is that more water is produced than the volume injected, namely, the water liquidation gets in the forefront of the field management. Since 1975 the Algyő field has been in the focus of different chemical EOR/IOR programs. Although an intensive study of different EOR methods addressing the whole reservoir space are still in the center of present efforts, the local stimulation of wells represents more attractive alternatives for the operator. Among others profile correction, water and gas shut-off, dewaxing, clay stabilization, etc. methods were implemented at industrial scale. In the present paper, however, only those methods are discussed in detail which are based on application of silicates.

Table 1.
Characteristics of the Algyő field

Depth, m	1900 - 2000
Thickness of pay zone, m	20 – 25
Porosity, fraction	0.15 – 0.25
Permeability, mD	100 – 500
Temperature, ^oC	92 – 95
Pressure, MPa	19.5
Rock type	sandstone
Oil type	light paraffinic
Water type	Na-hydrogen carbonate

The first comparative treatments of producing wells were performed during 1980-81 in the north-western section of the Algyő-2 reservoir. Because the combined polymer/silicate technique proved to be much better than the conventional polymer technology, extension of the project was decided. The routine application began in 1982 and bull head injection was used for all producers through a single perforation located near to the water/oil contact (WOC). Before the program ended in 1987, 16 producing wells were treated. In one case a repeated treatment was performed in 1984. The well selection was based on reservoir geological data and production history. Wells with a water cut exceeding 80 % were determined to be good candidates for treatment.

On average, 0.5 t polymer, 10.0 t SiO_2 containing alkali silicate, 0.4 t alum, and 0.2 t calcium chloride were used. The polymer had a high molecular weight ($9.5x10^6$ g/mol) and partially hydrolyzed (19 %) polyacrylamide. The polymer and the SiO_2 content in the treating solution were 2 g/dm^3 and 50 g/dm^3, respectively. The cross-linking solution contained 2 g/dm^3 alum and $CaCl_2$. HCl prevented early precipitation of aluminum hydroxide and controlled the pH in the mixing zone and the gelation of silicate. The surface facilities consisted of mobile elements, and the treating solutions were made at the well site. Technical failures and injectivity problems were seldom encountered. The solutions were injected into the wells at rate of 50 m^3/d and 20 to 40 bar on average, and maximum 20 bar increase in injection pressure was allowed during treatment. Deterioration of injectivity (from 1.0 to 2.5 m^3/d bar to 0.5 to 1.5 m^3/d bar) was moderate, and a minimum injection rate of 20 m^3/d could always be maintained.

The surplus oil production was calculated by comparing the statistical prediction of oil production through 6 months before treatment and the actual net output obtained after the treatment. The best field results were obtained in wells located in highly heterogeneous layers. It is interesting to note that the number of unsuccessful treatments in the second part of the project seems to be greater than in the first part. One probable reason could be the shift of the original WOC, which resulted in an unfavorable condition for treatments performed years later. However, the relatively large number of treatments and the time elapsed enable us to draw another remarkable conclusion: the systematic well treatments at the WOC have an advantageous impact on the production characteristics of untreated producing wells located in this section.

As the data in Fig. 3 illustrate, the incremental oil production totaled 95,000 t until the end of 1991. Although the statistical data showed only 50 % positive probability, the rate of return was greater than 40 and on average more than 5000 t of oil was produced by a single treatment. The overall budgetary balance of the project is definitely attractive and may accelerate the extension of the well treatments to other blocks and fields.

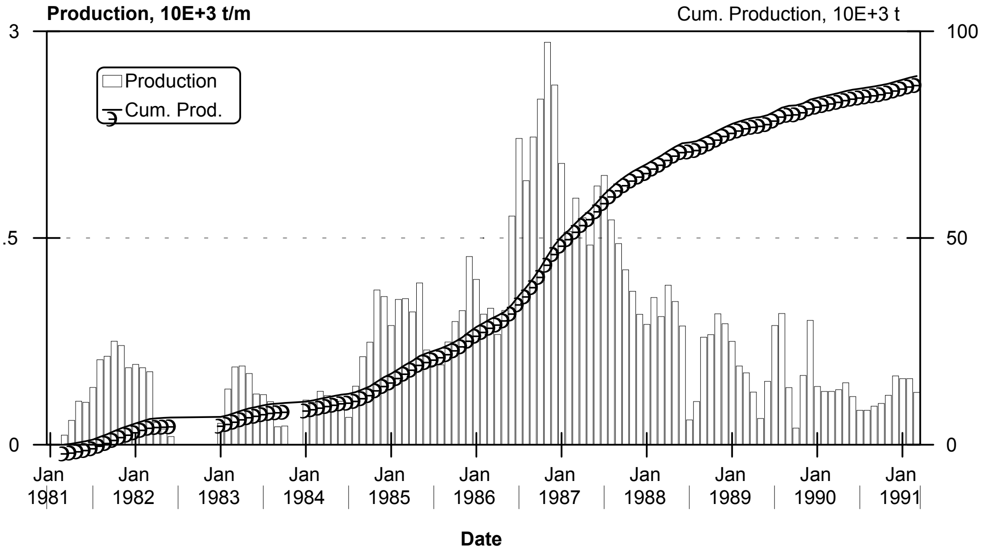

Fig. 3. Cumulative results of the polymer/silicate well treatment technique applied at the Algyő field between 1982 and 1988

In 1996 a new field project was started. This programme, consisting of 25 treatments was accomplished mainly in the north-western section of the same reservoir (Algyő-2), but five treatments were performed in an other hydrodynamic unit (Szőreg-1). All wells, except one, were producers. The chemical and the surface technology were nearly the same, but occasionally the type of polymer was different to maintain compatibility between the formation rock and the macromolecular material. In case of injector the consumption of chemicals, and hence, the penetration depth of treating solutions, were higher by 50 % than in case of producers.

At this time it is prematured to give a comprehensive analysis of the second project, however, it is probable that the overall performance will be less positive than it was in the previous project. More than five or ten years after the first project the reservoir system is more exhausted, the WOC and the position of the displacement front is more uncertain and at last but not least, the physical condition of wells are also worst. Recently, the available results and information notify only a marginal profit in advance.

Evaluating the well behavior after approximately forty treatments a subtle picture is outlined by the diverse production characteristics. The field experiences can be classified into the following groups:

1. The positive effect was long-lasting and the water cut decreased significantly (>20 %) immediately after the treatment. Afterwards, the water cut shown slow and gradual increase in time through many years (Fig. 4). This type of treatment is also characterized by substantial amount of oil production (>8000 t)
2. In certain cases the change of water cut was also measurable, its trend as expected (wave-like), but the positive effect was short (<1 year). As a result, the additional oil production was usually less than 1000 t (Fig. 5).
3. Some treatments yielded significant amount of surplus oil production even though sporadic change characterized the water cut (Fig. 6). Since the effect could be detected through many years, the cumulative oil production reached, or often exceeded 5000 t.

4. An outstanding situation was encountered in case of Alg.-234 well, which was treated two times. Response of well could be qualified as ideal as far the production characteristics are concerned (Fig. 7). The positive effect of the first treatment was negligible, but after the second one unusually great amount of additional oil production was registered. That case was already discussed in detail in a previous paper [16].
5. In a few cases the water cut decreased less than 5 %, however the oil production was stabilized for long time. Technically this might be considered as a positive result, but the operator qualified the job as non-profitable.
6. The share of unsuccessful treatments was about 30 %. In these cases the productivity often remained unchanged and the phase composition of the produced liquid was also constant or it kept its rising tendency in time.

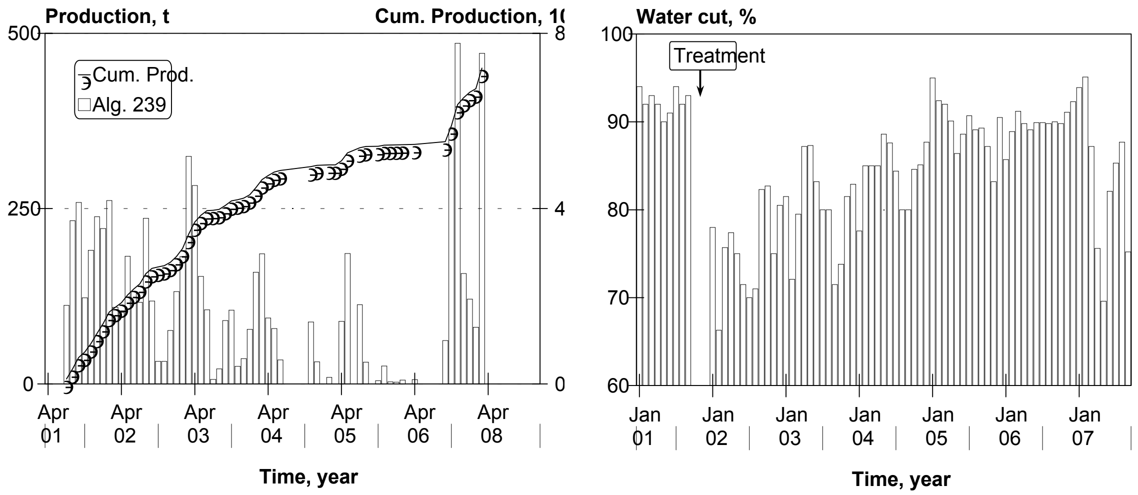

Fig. 4. Results of the polymer/silicate well treatment applied at the Alg. 239 well

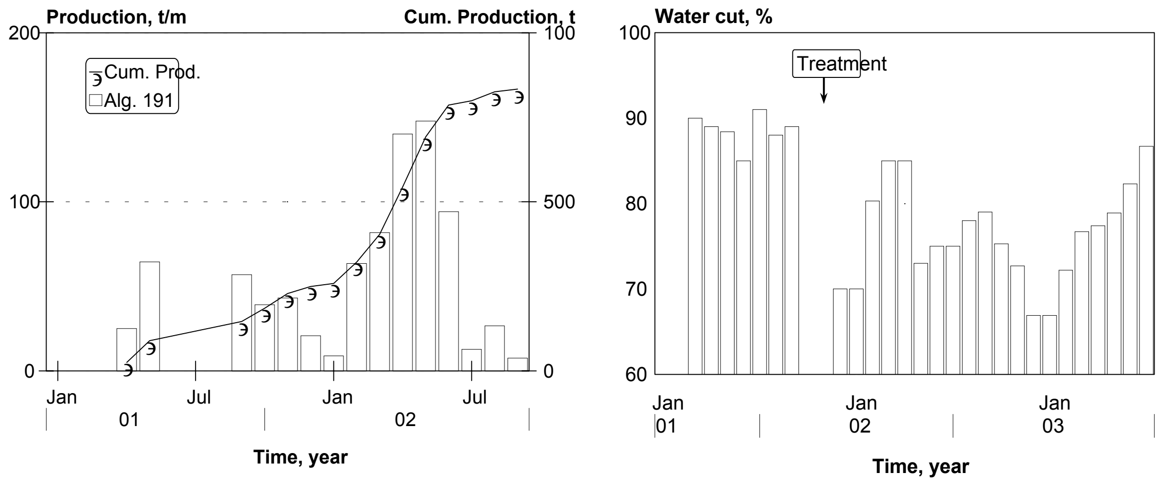

Fig. 5. Results of the polymer/silicate well treatment applied at the Alg. 191 well

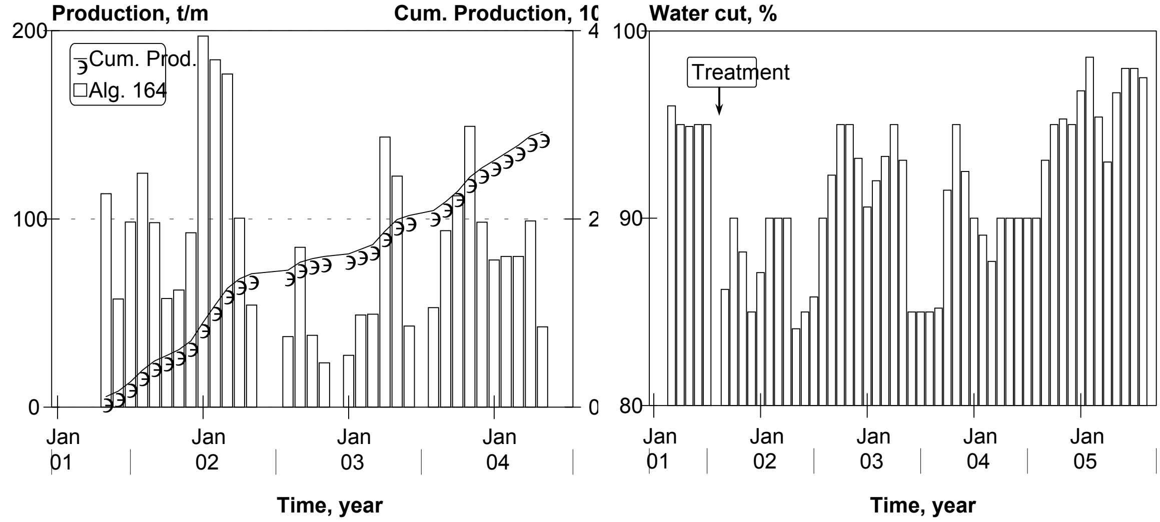

Fig. 6. Results of the polymer/silicate well treatment applied at the Alg. 164 well

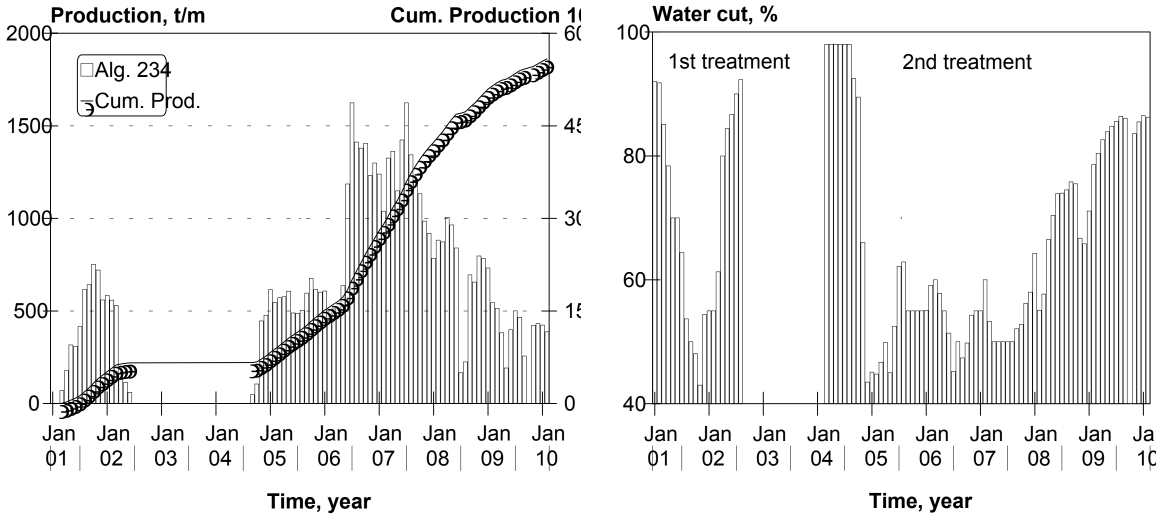

Fig. 7. Results of the polymer/silicate well treatment at the Alg. 234 well

These observations may serve as a basis for evaluation of the field projects. After all, it was concluded that sometimes the uniform displacing front already overrun the perforation, but sometimes the front was highly dispersed and responsible for entrapment of huge amount of mobile oil in the reservoir. Existence of fractures in the vicinity of treated wells was not likely. The best results were obtained in those cases when the vertical heterogeneity around the wells was extremely high and the hydrodynamic measurements proved a fair profile correcting effects. It was also demonstrated that under similar reservoir conditions a deeply penetrating (>20 m) placement technology has only chances for restriction of water production. The long-lasting effects, the radical changes and the high probability of positive effects unequivocally proved that the polymer/silicate method universally meet the practical expectation at the field.

CONCLUSIONS

1. By joint application of silicates and macromolecular materials a multifunctional, self-controlling chemical system is formed which works spontaneously even under harsh reservoir conditions.
2. The polymer/silicate method is inexpensive, flexible and adaptable to any production technology, meanwhile they eliminate most of the drawbacks of the original, pure silicate technologies.
3. It was shown that the silicates, combined with polymers offer unique opportunity to cure numerous production/injection problems (water-shut-off, profile correction, gas coning, etc).
4. Between 1980 and 1998 the field projects comprising more than forty well treatments yielded substantial additional oil production, life time of the wells were extended and the overall profitability of the field was significantly increased.
5. The Hungarian experts are convinced that the silicate-based methods are reasonable alternatives wherever and whenever the application of bulk and surface deposited gels is arising.

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