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Development of the semi empirical approach on wormhole formation in carbonates

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ABSTRACT

Carbonate acidizing is a technology of well productivity stimulation in oil recovery practice. The maximum effect of such a well treatment is connected with acidizing channels formation of several millimeters in diameter (wormholes). One of the approaches to predict the acidizing effect of wormholing is the semi empirical simulation based on the results of the experiments of core plug channeling by acid injection. It was established that maximum of permeability improvement corresponds to the minimum of injected volume and is defined by wormhole breakthrough. Presentation of such equation in the dimensionless form involves dimensionless criteria such as Damköhler, Peclet, etc, that are defined by the results of the calculation and with experiment data matching. The new approach to derivate of the main equation defining the acid wormholing process is developed in the present paper.

KEYWORDS

wormhole formation in carbonate core plug • dimensionless numbers by Damköhler and Peclet acidizing of carbonate reservoir

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Introduction

Well's productivity stimulation in carbonate reservoirs by acid solutions had a practice history from the middle of the previous century [1–3]. The main target of carbonate formation acidizing is the permeability improvement in the near well bore zone. Application of different technologies was proposed for this purpose: from low-rate acid injection for cavity creation near a well to high velocity injection that leads to reservoir fracturing and fracture stabilization by etching of its side surfaces. The ideology of channeling or wormholing of an oil formation was developed in the works [4–7].

A lot of the studies analyzed the formation and structure of wormholes in carbonates with the radioscopy and computer tomography of a core plug during acid injection [8,9]. In [10,11], it was used the large, massive carbonates blocks with a cylindrical channel or the well bore model. Application of unprecedented experimental scale helps to understand the formation of wormhole net in the vicinity of a well, their structure including fractal form.

These investigations helped to develop the general semi empirical theory of wormhole growth. It was established that the injection rate defines the following modes of the stimulation process: frontal acidizing of carbonate matrix and cavity creation near the inlet of core plug due to low-rate acid injection; wormhole growth with critical value of injection rate and volumetric acid reaction accompanied by high velocity acid flow. The maximum of well productivity increase occurs due to deep acid penetration to the matrix with formation of high permeability wormholes. The understanding of the mechanisms was achieved in the analysis of linear core plug experiments. The result of these studies established that wormhole formation is operated by injection rate through the Damköhler number or relation of reaction rate to acid flow velocity. The Peclet (or the ratio of convective to diffusive fluxes) and the acid capacity (defining the specific mass of matrix that can be dissolved by acid in pore space) numbers describe the influence of other parameters on the process. Its role in the reaction was considered in [12]. Note that the Peclet number is proportional to the injection rate but the Damköhler number is inverse proportional to the same parameter. Introducing the critical values of injection rate and volume of acid needed for wormhole breakthrough Fredd and Fogler [5] proposed new dimensionless parameters of the process: the ratio of injected acid volume and the rate to the critical values of these characteristics. Using these variables, they established the unique dependence: introduced specific acid volume from the Damköhler number. The physical meaning of the dependence is the connection of acid injection volume with the length of the created wormhole. Processing of the experiments by different investigators covered limestone and dolomite, various types of acid, their concentration, reaction and injection rates confirmed the unique dependence. The reservoir mineralogy of cause plays a definite role in the value of dimensionless numbers. Note that the heterogeneous reaction rate is defined from the experiments with the rotating disk apparatus [3] and by the volumetric method [13] and the definition of Damköhler number may involve internal wormhole parameters (radius, length) [4] and external characteristics (permeability, flow velocity) [14].

Acidizing experiments in a vuggy carbonate showed that specific acid volume to breakthrough is an order of magnitude lower than what has been observed with relatively homogeneous limestone [15]. Large-Scale Dual-Porosity Approach is developed for such reservoirs in [16].

The idea of the developing of the simulator including all acidizing modes attracts many investigators [8]. These approaches may be divided into several categories. Huang et al. [17] used the capillary tube approach to predict wormhole population density and calculate the volume of acid required to create wormholes of a certain density and length. Semi empirical models will be considered further in details. Wang et al. [18] developed transition pore theory to calculate the optimum flux to generate dominant wormholes during a matrix acid treatment. As a network model approach Fredd and Fogler [6] proposed the model to describe wormhole growth in 3D physically representative network where distribution of acid concentration is calculated due to transport and reaction with sphere grains. The last group uses Darcy scale models for acid transport in reacting matrix [19].

Further only semi empirical model group will be considered. This group [8] considers micromechanics of creation and growth of the main wormhole in a carbonate core plug. The underground of this model group is the laboratory investigation of wormhole creation and growth in a carbonate core plug and the study of heterogeneous reaction (in situ diffusion and reaction rate) on the rotating disk installation. As a result, these models involve the formulation of the main equation bonding pore volume of acid injection till its breakthrough to the outlet of core plug with the injection rate and also the influence of other parameters such as acid type and concentration [4], temperature [20] on the main equation. Note that it is difficult to find out the systematic study of reliable quality in the present studies, most researches give the 2–5 experimental points. Several investigators consider the main dimensionless characteristics of the process is the Damköhler number [6], other outline the Peclet number [20,21], Gong and El Rabaa [14] use mixed approach.

The impact of effective tortuosity of porous media of the Damkohler and Peclet dimensionless numbers is considered in [22]. Wang et.al analyzed the process of matrix acidizing in naturally fractured carbonate reservoirs [23]. The influence of acid type and combined acid systems on the process was studied experimentally in [24].

Summarizing the above considerations, the main equation bonding acid breakthrough pore volume (*PV*) and injection rate (*q*) has the following structure [14]:

$$PV = \frac{A}{q^2} + Bq^{\frac{1}{3}}, \tag{1}$$

where *A* and *B* are the constants depending on carbonate mineralogy, acid composition and concentration, in situ diffusion and some other factors. These constants are defined by matching calculation with the experimental data.

Gong and El Rabaa [14] integrated the approaches of Fredd and Fogler [4] and Daccord, Touboul and Lenormand [18] and proposed the following dimensionless version of the main Eq. (1). Their equation involves the Damköhler N_{Da}, the Peclet N_{Pe} and the acid capacity N_{ac}:

$$PV = f_1 \frac{N_{Da}}{N_{Pe}} + f_2 \frac{N_{Pe}^{\frac{1}{3}}}{N_{ac}}, \tag{2}$$

where f_1 and f_2 are the empirical constants defined from matching experimental data.

In [14], the dimensionless numbers are defined as the following:

$$N_{Pe} = \frac{q\sqrt{k}}{D} \quad N_{ac} = \frac{\emptyset \varkappa_R c \rho_w}{(1-\emptyset)\rho_R} \quad N_{Da} = \frac{D^{\frac{5}{3}}\sqrt{k}}{\nu^{\frac{2}{3}} q} \text{ (for dolomite)}, N_{Da} = \frac{K D^{\frac{2}{3}} k}{\nu^{\frac{2}{3}} q} \text{ (for limestone)}, \tag{3}$$

where *D* is the in-situ diffusion coefficient, *K* is the reaction kinetic constant, *k* and Ø are the permeability and porosity of a rock matrix, ρ_w/ρ_R is the ratio of acid solution and rock matrix densities, *ν* is the kinematic viscosity, $\varkappa_R$ is the solubility of matrix in acid, *c* is an acid concentration.

Further exactly these expressions of the dimensionless numbers will be used in the paper. Unfortunately, incorrect mathematical calculations hinder the application of the Gong and El Rabaa model for practical prediction of the process.

The main conclusion of the experimental studies is the establishment of the critical value of acid injection rate q_{cr} that accompanies wormhole formation and minimum pore volume injection till acid breakthrough PV_{cr}. The dependence of dimensionless variables $PV/PV_{cr} = f(q/q_{cr})$ is a universal function that is defined only by the Damköhler number.

Note that most of the models of other mentioned simulation approaches were verified by matching the calculations with the results of leaner experiments which are the framework the of semi empirical approach [25,26]. In a manner, the accuracy of these approaches is approximately the same as for semi-empirical models based on *PV* breakthrough curves from core scale experiments.

Formulation of the main theorems

Let's accept the main hypothesis about the dependence of breakthrough pore volume *PV* from the acid injection rate (1) by more general function:

$$PV = \frac{A}{q^m} + Bq^n. \quad (4)$$

The minimum of this function may be obtained by differentiating with respect to the injection rate and setting it to zero. The obtained equation may be rearranged to yield the expression of q_{cr}. Finally, the critical rate and the minimum/critical breakthrough pore volume are expressed as:

$$q_{cr} = \left(\frac{mA}{nB}\right)^{\frac{1}{m+n}}, \quad PV_{cr} = \frac{A^{\frac{n}{m+n}}B^{\frac{m}{m+n}}(m+n)}{m^{\frac{m}{m+n}}n^{\frac{n}{m+n}}}. \quad (5)$$

The first item. Let's prove the statement that Fredd's and Fogler's universal equation of wormhole growth is the consequence of the adoption of the equation in the form (1) and depends on only the values of exponents *m* and *n*. For this purpose, let's introduce the new dimensionless parameters PV/PV_{cr} and q/q_{cr} and rearrange the Eq. (4) to the following form:

$$P\frac{PV}{PV_{cr}} = \frac{1}{(m+n)}\left(n\left(\frac{q_{cr}}{q}\right)^m + m\left(\frac{q}{q_{cr}}\right)^n\right). \quad (6)$$

If we choose the inverse Damköhler number as the dimensionless injection rate than Eq. (6) will be defined by one universal constant *a*:

$$\frac{PV}{PV_{cr}} = \frac{1}{(m+n)}\left(n(aN_{Da})^m + \frac{m}{(aN_{Da})^n}\right). \quad (7)$$

Fredd and Fogler have determined this constant by matching calculations and experimental data and obtained the value 0.29. Consequently, the initial hypothesis (1) and (4) lead to the conclusion of the existence of universal dependence:

$$\frac{PV}{PV_{cr}} = f\left(\frac{1}{N_{Da}}\right). \quad (8)$$

The proved statement is true for various values of *m* and *n*.

Taking into account the dependence of introduced dimensionless numbers from injection rate (4) and Eq. (7) it is expedient to consider the following dimensionless numbers and their combination:

$$N_{ac};\ N_{Da};\ N_{FG} = N_{Pe}N_{Da}. \quad (9)$$

Here only Damköhler number depends on the injection rate that is why we can suppose that the critical value of parameters involve only N_{ac} and N_{FG} and breakthrough pore volume *PV* is defined by Eq. (4). The combination N_{FG} is applied in the theory of solutions separation by filters [27] and further will refer as the generalized Damköhler number. The authors propose the following expressions for the critical values:

$$PV_{cr} = \frac{g_1 {N_{FG}}^{\varepsilon}}{N_{ac}}, \qquad q_{cr} = g_2 {N_{FG}}^{\theta}(N_{Da}q), \quad (1)$$

where ε and θ are arbitrary exponents defined by matching experimental and calculated data. The proposed expressions (10) are based on the physical considerations that critical breakthrough pore volume PV_{cr} is directly bonded with matrix pore volume that could react with acid injected volume or $1/N_{ac}$.

The second item. The proposed approach bases on the main postulate that the wormhole growth mechanisms is defined by introduced critical values but the Eqs. (1) or (4) matches the injection rate only in the narrow rate interval. The extension of the dependence in the modes of frontal or volumetric acidizing or mixed reaction modes is not correct. The common trends of the main equation in the regions of high and low injection rates tends to infinity, from the other hand the objectives of frontal and volumetric reaction modes have definite solutions with the finite values. For example, in the low rate region, the breakthrough pore volume is defined by $1/N_{ac}$.

Let's analyze the experimental data on breakthrough pore volume, for example gathered in [6], which are presented on Fig. 1. First of all, the majority of experimental point lays in the vicinity of the critical injection rate. The number of points far from the critical value is less and the deviation of them is much higher. If we adopted the universal dependence in the form (4) than we can restrict the experimental points by two curves with exponents values: 1) $m = 2$, $n = 1/3$ and 2) $m = 5/2$, $n = 3/5$. In order to get away from the details the coordinates of points were normalized on Fig. 1. Therefore, the main target of the model's equation is to describe the behavior of PR function near the critical point and to define the critical parameters with sufficient accuracy.

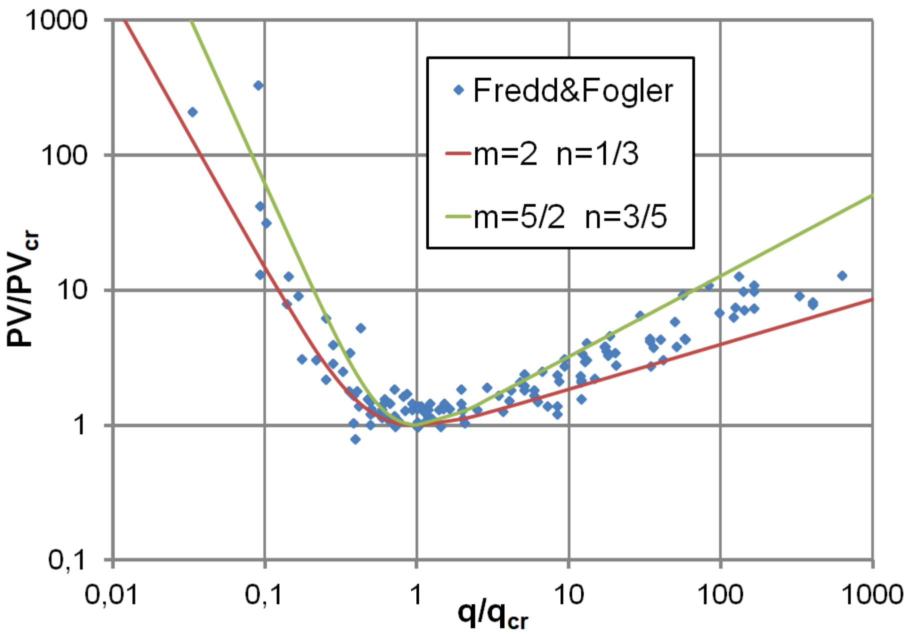

Fig. 1. Experimental data on normalized dependence of the breakthrough pore volume from dimensionless injection rate extracted from [6]

The third item. The proposed approach generalized the models presented by Fredd and Fogler [6], Gong and El Rabaa [14] and does not contradict the empirical results by Dong, Zhu and Hill [28], Pange, Ziauddin and Balakotaiah [29]. In the first item, it was established that the universal Fredd's and Fogler's equation (8) is defined by the assumptions about the structure of equations and could be arranged to the view (2) by simple mathematical calculations.

The main proposal of Gong and El Rabaa on the dimensionless numbers that operates the process is the same in the suggested approach. The equation (4) generalizes the Eq. (1) proposed by Gong and El Rabaa.

Dong, Zhu and Hill [28] analyzed experimental data executed by Wang et al. [18] and their own on temperature and acid concentration influence on the critical injection rate. They established the invariant that does not dependent from temperature and acid concentration. This invariant is defined as follows:

$$\frac{PV_{cr}D}{q_{cr}} = \text{const.} \tag{11}$$

After substitution the expressions of the critical parameters (10) to the invariant (11) one can obtain that the Dong's, Zhu's and Hill's conclusion is performed when ε and θ satisfy the condition: $\varepsilon = \theta + 1$.

This equation with Fredd and Fogler assumption lead to the following expression of critical parameters: $\varepsilon = 1,\quad \theta = 0,\quad PV_{cr} = \frac{g_1 N_{FG}}{N_{ac}},\quad q_{cr} = g_2(N_{Da}q),\quad g_2 = a = 0.29.$

If we take into account the proposal of Pange, Ziauddin end Balakotaiah on the equation form than we shall come to the following conclusions: $\varepsilon = 0.5,\quad \theta = -0.5,$ $PV_{cr} = \frac{g_1\sqrt{N_{FG}}}{N_{ac}},\quad q_{cr} = \frac{g_2(N_{Da}q)}{\sqrt{N_{FG}}}.$

At last, despite the mathematical claims to the work of Gong end El Rabaa we present the expressions of critical parameters in their model: $\varepsilon = \frac{1}{7},\quad \theta = -\frac{4}{7},$ $PV_{cr} = \frac{g_1(N_{ac}N_{FG})^{\frac{1}{7}}}{N_{ac}},\quad q_{cr} = \frac{g_2(N_{Da}q){N_{ac}}^{1/7}}{(N_{ac}N_{FG})^{4/7}}$ but instead of N_{FG} these expressions involve dimensionless constant N_{FG}, N_{ac} and critical injection rate includes excess N_{ac}.

Conclusions

Generalizing the conclusions of the proved items the following results can be declared:

1. The proposed approach integrates the main aspects of Fredd's and Fogler's theory on the influence of Damköhler number on the wormhole formation in carbonates, the proposal of Daccord, Gong et al. on the role of Damköhler, Peclet and acid capacity numbers in the acidizing process and the type of the equation bonding breakthrough pore volume with acid injection rate, established by Dong, Zhu end Hill invariant (10) is also being executed by Eq. (11).
2. The main purpose of the equation bonding breakthrough pore volume with acid injection rate is the precise determination of critical parameters. The key factors of the wormholing process are the critical parameter PV_{cr} and q_{cr}, which are dependent from Damkeler, Peclet and acid capacity numbers. The critical parameters do not dependent from exponents n and m, that is why the role of these parameters is limited by the approximation of breakthrough pore volume from injection rate and precise determination of critical parameters from experimental data.
3. The experiments on the influence of acid concentration, matrix permeability, in situ diffusion on critical parameters are of current interest. These experiments could help to select the most accurate models of wormholing growth in carbonates [30,31].

References

1. Pourabdollah K. Matrix acidizing: a fouling mitigation process in oil and gas wells. *Reviews in Chemical Engineering*. 2020;36(2): 311–331.
2. Krivoshchekov SN, Vyatkin KA, Ravelev KA, Kochnev AA. Influence of geological and technological parameters on effectiveness of hydrochloric acid treatment of carbonate reservoirs. *International Journal of Engineering*. 2020;33(10): 2113–2119.
3. Schechter RS. *Oil Well Stimulation*. Englewood Cliffs: Prentice Hall; 1993.
4. Fredd CN, Fogler HS. The kinetics of calcite dissolution in acetic acid solutions. *Chemical Engineering Science*. 1998;53(22): 3863–3874.
5. Fredd CN, Fogler HS. Influence of Transport and Reaction on Wormhole Formation in Porous Media. *AIChE Journal*. 1998;44(9): 1933–1949.
6. Fredd CN, Fogler HS. Optimum Conditions for Wormhole Formation in Carbonate Porous Media: Influence of Transport and Reaction. *SPE Journal*. 1999;4(03): 196–205.
7. Buijse MA. Mechanisms of Wormholing in Carbonate Acidizing. In: *Proceedings of the International Symposium on Oilfield Chemistry, 18–21 February 1997, Houston, Texas*. p.SPE–37283–MS.
8. Akanni OO, Nasr-El-Din HA. The Accuracy of Carbonate Matrix-Acidizing Models in Predicting Optimum Injection and Wormhole Propagation Rates. In: *Proceedings of the SPE Middle East Oil & Gas Show and Conference, 8–11 March 2015, Manama, Bahrain*. p.SPE–172575–MS.
9. Glasbergen G, Kalia N, Nalbot M. The Optimum Injection Rate for Wormhole Propagation: Myth or Reality. In: *Proceedings of the 8th European Formation Damage Conference, 27–29 May 2009, Scheveningen, The Netherlands. p*.SPE–121464–MS.
10. McDuff D, Shuchart CE, Jackson S, Postl D, Brown JS. Understanding Wormholes in Carbonates: Unprecedented Experimental Scale and 3-D Visualization. In: *Proceedings of the SPE Annual Technical Conference and Exhibition, 19–22 September 2010, Florence, Italy*. p.SPE–134379–MS.
11. McDuff D, Shuchart CE, Postl D. Understanding Wormholes in Carbonates: Unprecedented Experimental Scale and 3D Visualization. *J Pet Technol*. 2010;62(10): 78–81.
12. Pasha P, Taghinia F, Nadalinia Chari F, Jalili B, Jalili P, Domiri Ganji D. Chemical reaction-diffusion model around a vessel for studying temperature and concentration of three chemical species by finite element method. *International Journal of Engineering, Transactions A: Basics*. 2023; 36(1): 171–181.
13. Kremleva TA, Smirnov AS, Fedorov KM. Simulation of carbonate acidizing process including wormholing. *Izvestia RAN. Fluid mechanics*. 2011;5: 76–84. (In Russian)
14. Gong M, El-Rabaa AM. Quantitative Model of Wormholing Process in Carbonate Acidizing. In: *Proceedings of the SPE Mid-Continent Operations Symposium, 28–31 March 1999, Oklahoma City, Oklahoma*. p.SPE–52165–MS.
15. Izgec O, Zhu D, Hill AD. Numerical and experimental investigation of acid wormholing during acidization of vuggy carbonate rocks. *Journal of Petroleum Science and Engineering*. 2010;74(1–2): 51–66.
16. Cohen CE, Ding D, Bazin B, Quintard M. A New Matrix-Acidizing Simulator Based on a Large-Scale Dual-Porosity Approach. In: *European Formation Damage Conference, 30 May–01 June 2007, Scheveningen, The Netherlands*. p.SPE-107755-MS.
17. Huang T, Zhu D, Hill AD. Prediction of wormhole population density in carbonate matrix acidizing. In: *SPE European Formation Damage Conference, 31 May–01 June 1999, Hague, The Netherlands*. p.SPE-54723-MS.
18. Wang Y, Hill AD, Schechter RS. The Optimum Injection Rate for Matrix Acidizing of Carbonate Formations. In: *SPE Annual Technical Conference and Exhibition, 3–6 October 1993, Houston, Texas*. p.SPE-26578-MS.
19. Jia C, Sepehrnoori K, Huang Z, Jun Y. Modeling and Analysis of Carbonate Matrix Acidizing Using a New Two-Scale Continuum Model. *SPE J*. 2021;26(05): 2570–2599.
20. Daccord G, Touboul E, Lenormand R. Carbonate Acidizing: Toward a Quantitative Model of the Wormholing Phenomenon. *SPE Production Engineering*. 1989;4(01): 63–68.
21. Al-Arji H, Al-Azman A, Le-Hussain F, Regenauer-Lieb K. Acid stimulation in carbonates: A laboratory test of a wormhole model based on Damköhler and Péclet numbers. *Journal of Petroleum Science and Engineering*. 2021;203: 108593.
22. Alarji H, Alazman A, Regenauer-Lieb K. The impact of effective tortuosity on carbonate acidizing and the validation of Damköhler and Péclet dimensionless phase space. *Journal of Petroleum Science and Engineering*. 2022;212: 110313.
23. Wang J, Mo S, Zhao B, Liu Z, Tian X. Modeling matrix acidizing in naturally fractured carbonate reservoirs. *Journal of Petroleum Science and Engineering*. 2020;186: 106685.

24. Monette M, Nguyen QP. An Experimental Study of Wormhole Morphology with a Novel Acid System in Ultra-High-Temperature Carbonate Reservoirs. *Transport in Porous Media.* 2023;149: 625–642.
25. Ali M, Ziauddin M. Carbonate acidizing: A mechanistic model for wormhole growth in linear and radial flow. *Journal of Petroleum Science and Engineering.* 2020;186: 106776.
26. Ghommem M, Zhao W, Dyer S, Qiu X, Brady D. Carbonate acidizing: Modeling, analysis, and characterization of wormhole formation and propagation. *Journal of Petroleum Science and Engineering.* 2015;131: 18–33.
27. Oyama ST, Lim H. An operability level coefficient (OLC) as a useful tool for correlating the performance of membrane reactors. *Chemical Engineering Journal.* 2009; 151(1–3): 351–358.
28. Dong K, Zhu D, Hill AD. Theoretical and Experimental Study of Optimal Injection Rates in Carbonate Acidizing. *SPE J.* 2017; 22(03): 892–901.
29. Panga MK, Ziauddin M, Balakotaiah V. Two-scale continuum model for simulation of wormholes in carbonate acidization. *AIChE Journal.* 2005;51(12): 3231–3248.
30. Hoefner ML, Fogler HS. Pore evolution and channel formation during flow and reaction in porous media. *AIChE Journal.* 1988;34(1): 45–54.
31. Lucas CRDS, Neyra JR, Araujo EA, da Silva DNN, Lima MA, Ribeiro DAM, Aum PTP. Carbonate acidizing–A review on influencing parameters of wormholes formation. *Journal of Petroleum Science and Engineering.* 2023;220: 111168.

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