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Chapter

Tubular Heat Exchanger Fouling in Phosphoric Acid Concentration Process

Rania Jradi, Ali Fguiri, Christophe Marvillet and Mohamed Razak Jeday

Abstract

Fouling in phosphoric acid concentration is a persistent operational problem that compromises energy recovery in this process. Progress is hampered by the lack of quantitative knowledge of fouling dynamic effects on heat exchanger transfer. The object of this work is an experimental determination of the thermal fouling resistance in the tubular heat exchanger of phosphoric acid preheated installed in phosphoric acid concentration process. By measuring the inlet and outlet temperatures of phosphoric acid, steam temperature, suction and discharge pressure of the pump and acid density measurement, the overall heat transfer coefficient has been determined. The determination of the overall heat transfer coefficient with clean and fouled surfaces, allowed calculating the fouling resistance. The results from the heat exchanger studies showed that the fouling resistance increased with time and presented an asymptotic evolution in compliant with the proposed model by Kern and Seaton, with the existence of fluctuation. The poorly cleaned heat exchanger implied the absence of the induction period and caused, consequently, high values of the fouling resistance in a relatively short-time period.

Keywords: fouling, heat exchanger, heat transfer coefficient, fouling resistance, phosphoric acid concentration unit

1. Introduction

The main issues which various industries are facing are the accumulation of undesired substances or materials dissolved or presented as a suspension in the fluid on the heat transfer surfaces [1]. This phenomenon which is called as fouling affects the equipment operation by reducing their thermal effectiveness. This causes a significant economic loss due to the installations of regular cleaning [2, 3].

Fouling in heat transfer process is often inevitable and reduces energy efficiency and plant operability. Mitigation of fouling, and effective cleaning strategies, both require understanding the mechanisms involved in deposition and cleaning [4]. Many researches on fouling in heat transfer processes are dealt with, by reducing the efficiency of heat transfer and limiting productivity [5]. Phosphoric acid fouling in concentration process preheat exchangers is a persistent operational problem that

compromises energy recovery in these process. Progress is hampered by the lack of quantitative knowledge of the dynamic effects of fouling on heat transfer exchanger [6]. Generally, phosphoric acid, which is the cold fluid, flows through the tube side while steam, which is the hot stream, flow through the shell side in heat exchangers [7]. The solution of concentrated phosphoric acid is supersaturated with calcium sulfate, resulting in the deposition on the contact material [8]. Given that the thermal conductivity of these scales is low, even a thin layer of scale can drastically reduce the overall heat transfer coefficient [9]. Furthermore, fluorosilicate and fluoroaluminate deposits on the acid ducts of clarifier tanks and evaporators can be imbedded in gypsum scale, which reduces pipe diameter and flow rate. In spite of considerable research efforts at the phosphoric acid type scale, no viable commercial solution has been found [10–12]. Behbahani et al. [13] have done a high number of fouling experiments in a side-stream of a phosphoric acid plant for various flow velocities, surface temperatures and concentrations in order to determine the mechanisms which control the deposition process. After identifying the effects of operational parameters on the deposition process, a fouling kinetic model by crystallization has been developed in Behbahani et al. [8]. A mathematical model has been elaborated to predict the fouling resistance in concentrating phosphoric acid [14]. The predicted fouling resistances were compared with the experimental data. Majority engineering calculations on heat transfer use the experimental heat transfer coefficients [15].

In this survey, we will examine the fouling phenomenon of the heat exchanger tubes for the preheat circuit of the phosphoric acid. The heat exchanger used for heating phosphoric acid is exposed to the fouling problem at the tube side of heat exchangers. In this context an experimental determination of the thermal fouling resistance by measuring the inlet and outlet temperatures of phosphoric acid, the temperature of steam, suction and discharge pressure of the pump and acid density measurement, the overall heat transfer coefficient has been determined. The determination of the overall heat transfer coefficient for the heat exchanger with clean and fouled surfaces makes it possible to calculate the fouling resistance.

2. Fouling mechanisms

Fouling can be divided into a number of distinct mechanisms. In general, many of these fouling mechanisms occur at the same time and each requires a different prevention technique. Among these different mechanisms, some represent different stages of the fouling process. The main mechanisms or stages of fouling include:

- 1. Period of initiation or delay. This is the clean surface period before dirt accumulation. This accumulation of relatively small deposits can even improve heat transfer over a clean surface and give the appearance of a "negative" fouling rate and a total negative fouling amount.
- 2. Particle fouling and formation, aggregation and flocculation.
- 3. Mass transport and migration of fouling agents to fouling sites.
- 4. Separation and deposition phase involving nucleation or initiation of fouling sites and attachment leading to deposit formation.
- 5. Growth, aging and hardening and increase of deposit resistance or auto-retardation, erosion and elimination.

3. Rate of fouling

Fouling is defined as a phenomenon that occurs with or without a temperature gradient in many natural, domestic and industrial processes. A surface is "dirty" when unwanted material accumulates there.

The fouling rate is normally defined as the average deposit surface loading per unit of surface area in a unit of time. Depending on the fouling mechanism and conditions, the fouling rate may be linear, falling, asymptotic or saw-tooth, as the case may be. **Figures 1** and **2** shows the different types of fouling rate.

1. Linear fouling is the type of fouling where the rate of fouling can be stable over time with the increase of fouling resistance and deposit thickness. It usually occurs when the temperature of the deposition in contact with the flowing fluid remains constant.

Ebert and Panchal [16] presented a fouling model expressing the average (linear) fouling rate under given conditions following two competing terms, namely a deposit term and an attenuated term.

Fouling rate = (deposit term) - (anti - deposit term)

$$\frac{dRf}{dt} = \alpha \ Re^{\beta} \ Pr^{\delta} \exp\left(\frac{-E}{RT_{film}}\right) - \gamma \tau_w \tag{1}$$



Figure 2. *Practical fouling curve.*

where α , β , γ and δ are parameters determined by regression, τ_w is the shear stress at the tube wall and T_{film} is the temperature of the fluid film (average of local bulk and local wall fluid temperatures). The relationship in Eq. (1) indicates the possibility of identifying combinations of temperature and velocity below where fouling rates will be negligible. Ebert and Panchal [16] present this as the "threshold condition." The model in Eq. (1) suggests that the geometry of the heat exchanger which affects the surface and film temperatures, velocities and shear stresses can be effectively applied to maintain the conditions below "threshold conditions" in a given heat exchanger.

- 1. Falling fouling is the type of fouling where the fouling rate decreases with time, and the deposit thickness does not reach a constant value, although the fouling rate never drops below a certain minimum value. In general falling fouling is due to an increase of removal rate with time. Its progress can often be described by two numbers: the initial fouling rate and the fouling rate after a long period of time.
- 2. Asymptotic fouling rate is where rate decreases with time until it becomes negligible after a period of time when the deposition rate becomes equal to the deposit removal rate and the deposit thickness remains constant. This type of fouling generally occurs where the tube surface temperature remains constant while the temperature of the flowing fluid drops as a result of increased resistance of fouling material to heat transfer. Asymptotic fouling may also result from soft or poorly adherent suspended solid deposits upon heat transfer surfaces in areas of fast flow where they do not adhere strongly to the surface with the result that the thicker the deposit becomes, the more likely it is to wash off in patches and thus achieve some average asymptotic value over a period of time. The asymptotic fouling resistance increases with increasing particle concentration and decreasing fluid bulk temperature, flow velocity, and particle diameter. The asymptotic fouling model was first described by Kern and Seaton [17]. In this model, the competing fouling mechanisms result in asymptotic fouling resistance beyond which any additional increase in fouling does not happen.
- 3. Saw-tooth fouling occurs where part of the deposit is detached after a critical residence time or once a critical deposit thickness has been reached. The fouling layer then builds up and breaks off again. This periodic variation could be due to pressure pulses, scaling, trapping of air inside the surface deposits during shutdowns or other reasons. It often corresponds to the moments of system shutdowns, startups or other transients during operation.

4. Fouling measurement and monitoring

The fouling resistances can be measured experimentally or analytically. The main measurement methods include:

1. Direct weighing: the simplest method for assessing the extent of deposition on laboratory test surfaces is to weigh directly. The method requires an exact balance in order to be able to detect relatively small changes in the mass of deposits. It may be necessary to use thin walled tube to reduce the tare mass in order to increase the accuracy of the method.

- 2. Thickness measurement: in many examples of fouling the thickness of the deposit is relatively small, perhaps less than 50 μm, so that a direct measurement is not easy to obtain. A relatively simple technique provided there is reasonable access to the deposit, consists in measuring the thickness. By using a removable coupon or plate, the thickness of a hard deposit such as a scale, can be obtained using a micrometer or traveling microscope. For a deformable deposit containing a large proportion of water, e.g., a biofilm it is possible to use an electrical conductivity technique.
- 3. Heat transfer measurements: in this method, the fouling resistance can be determined according to the changes in heat transfer during the deposition process. The equation for the following operations will be Eq. (11). The data can be reported in terms of changes in overall heat transfer coefficient. A major hypothesis of this method is that the presence of the deposit does not affect the hydrodynamics of the flowing fluid. However, during the first stages of deposition, the surface of the deposit is generally rougher than the metal surface so that the turbulence in the fluid is greater than when it is flowing on a smooth surface. As a result the fouling resistance calculated from the data will be lower than if the increased turbulence level had been taken into account. It is possible that the increased turbulence offsets the thermal resistance of the deposit and negative values of thermal resistance will be calculated.
- 4. Pressure drop: as an alternative to direct heat transfer measurements it is possible to use changes in pressure drop caused by the presence of the deposit. The pressure drop is increased for a given flow rate due to the reduced flow area in the fouled condition and the roughness of the deposit. The shape of the curve relating pressure drop with time will generally, follow an asymptotic shape so that the time to achieve asymptotic fouling resistance can be determined. The method is often associated with the direct measurement of thickness of the deposit layer. Friction factor changes can also be used to indicate fouling of a flow channel.
- 5. Other techniques for assessing fouling: with regard to their effect on heat exchanger performance the measurement of heat transfer reduction or increase in pressure drop provide a direct indication. The simple methods of measuring deposit thickness described above are useful, but in general they require that the experiment be completed in order to allow access to the test sections. Ideally non-intrusive techniques would allow further deposition while maintaining experimental conditions without disturbance. Such techniques include the use of radioactive tracers and optical methods. Laser techniques can be used to study the accumulation and removal of deposits. In addition, infra-red systems are used to study the development and removal of biofilms from tubular test sections. Microscopic examination of deposits may provide further evidence of the mechanisms of fouling, but this is usually a "backup" system rather than providing quantitative data.

5. Prediction of fouling factor

As noted above, fouling has the effect of forming on the heat transfer surface a substantially solid deposit of low thermal conductivity, through which heat is to be transferred by conduction. But as the thermal conductivity of the fouling layer and its thickness are not generally known, the only possible solution to the heat transfer problem is to introduce a fouling factor to take into account the additional resistance to heat transfer and possible calculation of the overall coefficient of heat transfer. A fouling coefficient is also sometimes specified, it is the reciprocal value of the fouling factor. When carrying out heat transfer calculations, the selection of fouling factors must be made with caution, especially when the fouling resistances completely dominate the thermal design.

The influence of inherent uncertainties in fouling factors is generally greater than that of uncertainties in other design parameters such as fluid properties, flow rates and temperatures [18]. An important fouling factor is sometimes adopted as a safety margin to cover uncertainties on the properties of fluids and even in the knowledge of the process, but the use of an excessively large fouling factor will result in an oversized heat exchanger with two or three times more area than is necessary. Although many tabulations based on the experiment are available and provide typical fouling factors such as the TEMA RGP-T-2.4 table [19], an acceptable assessment of the effects of fouling needs to be judged and evaluated for each particular application. Such tabulations can, however, serve as a guide in the absence of more specific information.

A number of semi empirical models have been proposed over the years for the prediction of the rate of fouling in heat exchangers or for estimating a fouling factor to be used in heat transfer calculations.

The first work on this subject began in the late 1950s with Kern and Seaton [17].

The modeling resulting from this work is based on the assumption that two processes act simultaneously. The first process is that of particle deposition characterized by a deposition flux that is constant if the concentration is also constant. The second process is that of the re-entrainment of particles characterized by a reentrain flow ϕ_r dependent on the mass of particles (m_p) deposited. The particle balance of the deposit is expressed according to the following equation:

$$\frac{dm_p}{dt} = \phi_d - \phi_r \tag{2}$$

The deposition process is designed as the serialization of particle transport and adhesion mechanisms. The following assumptions are made:

- consideration of a single type of fouling;
- homogeneity of the deposit;
- not taken into account of the phase of initiation of the deposit and the state of surface;
- constancy of the properties and thermo-physical characteristics of the fluid and the deposit.

The particle wall transport phase controls the deposition process while the shear stress controls the re-entrain phase of the particles. Thus, considering the proportionality of ϕ_d as a function of the deposited mass of particles, we can write the following equations:

$$\phi_d = k_p * (C_b - C_w) \tag{3}$$

$$\phi_r = C_1 * \tau_w * m_p \tag{4}$$

Or

 k_p is the transport coefficient.

 C_b the particle concentration in the fluid.

 C_w the particle concentration at the wall.

 C_1 a dimensional constant.

 τ_w the shear stress exerted by the fluid on the deposit.

Equation (2) thus becomes:

$$\frac{dm_p}{dt} = k_p * (C_b - C_w) - C_1 * \tau_w * m_p$$
(5)
The solution of Eq. (5) is thus:
$$m_p = \frac{K_p * (C_b - C_w)}{C_1 * \tau_w} * [1 - \exp(-C_1 * \tau_w * t)]$$
(6)

Assuming that $\tau = \frac{1}{C_1 * \tau_w}$ and $m_p^* = \frac{K_p * (C_b - C_w)}{C_1 * \tau_w}$. We can thus express the equation as follows:

$$m_p = m_p^* * \left[1 - exp\left(-\frac{t}{\tau} \right) \right] \tag{7}$$

Considering that the initial fouling flow is equal to the deposition flow and that the thermo physical properties of the deposit (conductivity and density) are constant, it is thus possible to express Eq. (7) in the form of a thermal fouling resistance:

$$Rf(t) = Rf^* * \left[1 - exp\left(-\frac{t}{\tau}\right)\right]$$
(8)

With Rf(t), the evolution of the fouling resistance as a function of time expressed in $[m^2-K/W] Rf^*$, the asymptotic value of the fouling resistance expressed in $[m^2-K/W]$ (this value characterizes the situation where the deposition rate equals the breakout speed). *t*, the time expressed in $[s] \tau$, the characteristic time expressed in [s] and whose value is generally attributed to the time required for the fouling resistance to reach its asymptotic value if the evolution of this kinetics was linear.

The Kern and Seaton [17] model therefore provides a mathematical description of the concept of simple fouling. This equation verifies the asymptotic behavior of the formation of a particulate deposit on the exchange surface of a heat exchanger. All models and theory of fouling are based on this model.

An apparent weakness of the Kern and Seaton [17] model is that the re-entrain flow depends on the thickness of the deposition layer. As a result, it is only once a significant deposit thickness has accumulated that the role of the re-entrain term becomes significant [20]. In the case of high speed flow, the deposit would be completely removed.

We also note that this model requires to go back on the values of Rf^{t} and τ . In general, there is no way to predict these values unless detailed experimental work has been done [21]. These values are thus established on a given installation and especially under given operating conditions. Thus, a modification, even minor, of the operating conditions (for example, water quality, flow modification) significantly modifies the parameters of the model and leads to a bad modeling of the fouling [22].

However, we note various works that make it possible to know the impact of the flow velocity (Um) on the value of the asymptotic fouling resistance Rf^* , or even to determine the value Rf^* .

Different authors thus propose a relationship of proportionality of type:

$$Rf^* \sim Um_i$$
 (9)

With regard to the tube exchangers: for Kern and Seaton [17] the value of *i* is -1. For Watkinson [23], this constant takes the value of -1.2 to -2.

As far as plate heat exchangers are concerned, Muller-Steinhagen [24] has in its study demonstrated a relation of proportionality between the asymptotic resistance of fouling Rf^* and the inverse of the speed squared (i.e., an exponent i - 2 in Eq. (9)), without providing a general relationship.

In the same context, Grandgeorge [25] proposes an empirical relation resulting from several experiments on different industrial size plate heat exchangers. In this context, Grandgeorge [25] established that the use of the initial pressure drop in the heat exchanger (ΔPo) in place of the flow velocity makes it possible to correlate with the aid of a single relationship the value of the asymptotic resistance Rf^* . The relationship is as follows:

$$Rf^* = \frac{1}{4 * \Delta Po} \tag{10}$$

Based on these observations, this model has been revised and modified by various researchers with various descriptions of the term deposition and re-entrain: Only empirical parameters were added and derived solely from the experimental study [20, 25].

6. Experimental procedure

The phosphoric acid concentration loop is allowed to concentrate—by evaporation—the phosphoric acid from 28 to 54% P_2O_5 in a forced-circulation evaporator closed loop, functioning under vacuum feeded by a barometric condenser. The system used for concentration composed of a stainless steel tubular heat exchanger, a centrifugal pump, a boiler or expansion chamber, a barometric condenser and a basket filter [26].

The inclusion of the dilute acid is done at the basket filter where it mixes with the circulating acid in order to protect the pump from abrasion and to limit the heat exchanger fouling, which reduces the stop frequency for washing. The circulation pump then aspirates the mixture formed and sends it to the inlet of the heat exchanger at a temperature in the order of 70°C. The heat exchanger allows heating the phosphoric acid at a temperature in the order of 80°C. The steam undergoes a condensation at a temperature of around 120°C at the level of the exchanger. The condensate will be sent to a storage tank before being returned to the utility center.

The overheated mixture of the acid outgoing the exchanger then passes into the boiler where an amount of water evaporates and the production of concentrated acid is done by overflow in inner tube of the boiler and the rest will be recycled. The condenser also ensures the re-entrain of incondensable outgoing of the boiler by the effect of water tube created by the waterfall. At the foot of the barometric guard, the seawater is gathered in a guard tank before being rejected to the sea (**Figure 3**).

Our experimental study is based on the following hypotheses.

- 1. The flow of two fluids (Phosphoric acid and steam) is at counter current.
- 2. Values of the thermo-physical properties of the fluids were considered constant.



Figure 3.

Simplified diagram of the phosphoric acid concentration unit.

3. The thermal losses were neglected.

- 4. The inlet and outlet temperatures of the two fluids are determined at the extremities of the heat exchanger.
- 5. Pump suction and discharge pressure measurements are taken at the extremities of the circulation pump.

7. Calculation method

The experimental data was collected out during 1 year. The method that we used to follow the fouling evolution consists in carrying out a heat balance at the boundaries of the heat exchanger by the intermediary of measurements of the inlet and outlet temperatures pump suction and discharge pressure measurements and acid density measurement (**Figure 4**). The latter was taken each 2 h during all the day.

This method, albeit indirect, makes it possible to detect the necessary moment to shut down the installation for cleaning. In the current study, the temporal evolution of the fouling resistance of the phosphoric acid was studied.

The calculation of the fouling resistance was done using the following relation:

$$Rf(t) = \frac{1}{U_{\rm s}} - \frac{1}{U_{\rm p}} = \frac{1}{U(t)} - \frac{1}{U(t=0)}$$
(11)

7.1 Calculation of U_s

The overall heat transfer coefficient at the dirty state was given in the time course, via the expression:



The measurement method at the boundaries of the heat exchanger.

$$U_s = U(t) = \frac{\dot{m}_{ac,cir} * Cp_{ac} * (T_{out,ac} - T_{in,ac})}{A * F * \Delta T_{ml}}$$
(12)

This relation is taken by the evaluation of energy on the heat exchanger by supposing the isolated system and the physical properties of the two fluids, as well as, the heat transfer coefficients stay constant along the exchanger.

7.2 Calculation of U_p

In the phosphoric acid concentration unit, the operating conditions at the limits of the heat exchanger unstable, it is necessary to disclose the heat exchange coefficients in the proper conditions U_p corresponding to the new operating conditions. Assuming that the cleaning between operational runs is perfect and that the heat exchangers are totally free of fouling at the beginning of a new run. The initial value of the overall heat transfer coefficient at the beginning of every cycle is considered as the value of the overall heat transfer coefficient in the clean state.

$$U_{p} = U(t = 0) = \frac{\dot{m}_{ac,cir} * Cp_{ac} * (T_{out,ac} - T_{in,ac})}{A * F * \Delta T_{ml}}$$
(13)

8. Results and discussion

8.1 Temporal evolution of the fouling resistance

The evolution of the fouling resistance in the phosphoric acid concentration process heat exchanger was followed for a study period quoted previously. This heat exchanger is already in service for 100 days before the be-ginning of the present study. However, it has carried out a stop that lasted 12 hours then its return to service. This stop is for the heat exchangers cleaning.

All the results from the fouling resistance are presented on **Figure 5**.

According to the values of these resistances, which are the majority higher than zero, this experimental data presents a fouling state. This is evident since, as mentioned before, this exchanger is in service for more than 3 months. The curves presented show that the temporal evolution of the fouling resistance, seems to follow an asymptotic evolution, which conforms to the model of Kern and Seaton [17], with the absence of the induction period. That can be explained to the rapid evolution of this phenomenon associated in particular with the characteristics of the



Figure 5.

Variation of the fouling resistance as a function of time for the stainless-steel-tubular heat exchanger.

treated phosphoric acid. As it appears clearly as the fouling resistance increases with the time until reaching a maximum value varied from 1.38×10^{-4} to $1.61 \times 10^{-4} \text{ m}^2$.K.W⁻¹.

The series functioned for more than 5 days, a sufficient period so that the resistance asymptotic value is reached. The fluctuation observed on these curves are due to the variation of flow, which, acting on the shear stress to the wall, causes re-entrain deposit particles or their deposition according to the sent flow value.

8.2 Temporal evolution of the overall heat transfer coefficient

From Eq. (11), we notice that the overall heat transfer coefficient is inversely proportional to the fouling resistance.



Figure 6.

Variation of the overall heat transfer coefficient as a function of time for the stainless-steel-tubular heat exchanger.



Figure 7. *Kinetics of fouling of the stainless-steel-tubular heat exchanger.*

The fouling resistance increases over time, which leads to a decrease in the flow of heat exchanged between the phosphoric acid and the steam, and subsequently the decrease in the overall heat transfer coefficient. As it appears clearly on **Figures 5** and **6**, when the fouling resistance increases with the time, the overall heat transfer coefficient decreases until reaching a minimum value varied from 1821 to 2078 W.m⁻².K⁻¹.

8.3 Temporal evolution of the fouling resistance obtained from both measurement and the Kern and Seaton model

One of the earliest correlative models for the characterization of the asymptotic kinetics of fouling, we distinguish Kern and Seaton [17], whose general expression is as follows:



This model gives rather satisfactory results, provided that the asymptotic value of the thermal resistance Rf^* as well as the time constant τ are evaluated, which strongly conditions the accuracy of the model.

The analysis of the experimental data which makes it possible to carry out the plots of **Figure 7** gives us the results of the two greatness Rf^* and τ for the stainless steel tubular heat exchanger. The asymptotic model is fairly faithful to the experimental data with determination coefficient R^2 close to 1 (**Table 1**).

$Rf^{*}[m^{2}.K.W^{-1}]$	τ [h]	R ²
$1.72^{*}10^{-4}$	40.32	0.975

Table 1.

Values of the asymptotic fouling resistance, the time constant and the determination coefficient for the stainless-steel tubular heat exchanger.

9. Conclusion

The monitoring of heat exchangers provides the sound knowledge of the fouling evolution in the specific conditions of the process. Deposit formation is a thermal resistance which leads important economic penalties.

The aim of this work was the study of the heat exchanger fouling phenomenon in the concentration phosphoric acid process. Secondly, the study concerned the temporal evolution of the fouling resistance and the overall heat transfer coefficient.

The results achieved revealed an asymptotic evolution of the fouling resistance, compliant with the model of Kern and Seaton with the lack of the induction period, which is explained by the consequence of an improper cleaning, or a deviation between the present study and the beginning of the heat exchangers functioning after the last stop.

Nomenclature

А	area, m ²
Ср	specific heat capacity, J.Kg ⁻¹ .K ⁻¹
F	correction factor (=1 for a steam condenser)
ṁ	mass flow rate, kg.s ⁻¹
Р	pressure, bar
Q	thermal power, W
Rf	fouling resistance, m ² .K.W ⁻¹
Т	temperature, K
t	time, h
U	overall heat transfer coefficient, $W.m^{-2}.K^{-1}$
Greek letters	
Δ	difference of greatness between two points
τ	time required to reach 63.2% of Rf^* , h
Subscripts	-
ac	acid
cir	circulation
ex	exchange
in reaction of the	input
ml	logarithmic mean
0	clean
out	output
р	clean state
S	dirty state
st	steam
*	asymptotic value

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