Effect of some organic surfactants on the corrosion behavior of low-carbon steel in hydrochloric acid solution*

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Abstract

The corrosion of 08PS mild steel in 2 M HCl in the presence of cationic (alkylbenzyldimethylammonium chloride (Catamin AB)), amphoteric (cocamidopropyl betaine (CAPB)), anionic (sodium lauroyl sarcosinate (SLS), sodium oleoyl sarcosinate (SOS), sodium dodecylbenzenesulfonate (SDBS)), and non-ionic surfactants (indirect material OP-10) was studied. The protective effects of the surfactants on steel corrosion decrease in the series: Catamin AB > CAPB > SOS > OP-10 > SLS > SDBS. The possibility of creating mixed corrosion inhibitors (CIs) based on the studied surfactants and urotropine, a known corrosion inhibitor of steel, is considered. It is shown that the Catamin AB + urotropine and CAPB + urotropine mixtures are the most promising formulations for the creation of efficient CIs of steel. These mixtures of CIs are recommended for application in HCl solutions with temperatures up to 60 and 80°C, respectively. The total content of the mixed CIs in the corrosive medium should be 5 mM. The molar ratio of a surfactant and urotropine in the mixed CIs is 1:9.

Received: July 25, 2023. Published: August 16, 2023 doi: 10.17675/2305-6894-2023-12-3-15

Keywords: *acid corrosion, steel, corrosion inhibitors, hydrochloric acid, alkylbenzyldimethylammonium chloride, cocamidopropyl betaine, urotropine.*

Introduction

Hydrochloric acid solutions are widely used at modern plants to remove various mineral formations (thermal scale, rust, salt deposits) from the surface of steel equipment and products. Corrosion inhibitors (CIs) are used to protect steel items in contact with HCl solutions. There is a wide range of compounds recommended for the protection of steels in HCl solutions [1, 2], but not many of these can be used as industrial CIs. In the production of industrial CIs, it is important that they be made from available and cheap raw materials that are produced on a continuous basis. In periods of instability of the world economic

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*This study was carried out as part of R&D (2022–2024): "Chemical resistance of materials, protection of metals and other materials from corrosion and oxidation" (EGISU registration number 122011300078-1, inventory number FFZS-2022-0013).

system, the production of many chemical compounds and their export to the world markets may decrease or stop completely. In such circumstances, it is necessary to search for compounds that would be potentially suitable as raw materials in the production of CIs. Previously, we studied medical preparations of the triphenylmethane series (brilliant green, malachite green) produced in large amounts as such a promising group of compounds [3].

Surfactants are an important group of industrial products widely represented on the world market [4–10]. The structure of surfactants includes hydrophobic and hydrophilic groups. Depending on the structure of the hydrophobic group, they are classified as:

- *Anionic.* The surface active part of the molecule carries a negative charge, *e.g.*, $[H_3C–(CH_2)_{11}–COO^-]Na^+$ has a long chain hydrophobic carrying the negative charge [11].
- *Non-ionic.* The surface active part of the molecule apparently carries no charge, *e.g.*, $H_3C–(CH_2)_{11}–O–(CH_2CH_2O)_7–H$ [12].
- *Cationic.* The surface active part of the molecule carries a positive charge, *e.g.*, $[H_3C–(CH_2)_{11}–N^+(CH_3)_3]Cl^-$ [13].
- *Amphoteric (Zwitterionic).* The surface active part of the molecule can carry a positive or negative charge, or both, depending upon the conditions, *e.g.*, $H_3C–(CH_2)_{11}–N^+(CH_3)_2–CH_2–COO^-$ [14].

Moreover, speciality [15] and polymeric surfactants [16] are distinguished. Surfactants with various structures are widely used as CIs of metals in corrosive media [17–20].

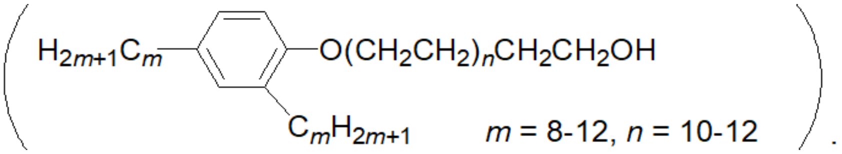
Table 1. Effect of surfactants ($C_{in} = 5$ mM) on the corrosion rate of 08PS steel in 2 M HCl (k) and the corrosion inhibition factors (γ). $t = 60$°C.

No.	**Surfactant**	**Label**	**k, g·m^{-2}·h^{-1}**	**γ***
1	Cocamidopropyl betaine	CAPB	8.4	13
2	Alkylbenzyldimethylammonium chloride	Catamin AB	3.1	34
3	Sodium lauroyl sarcosinate	SLS	25	4.2
4	Sodium oleoyl sarcosinate	SOS	17	6.2
5	Sodium dodecylbenzenesulfonate	SDBS	59	1.8
6	Indirect material OP-10	OP-10	23	4.6
7	Urotropine	–	10	11

* The corrosion rate of 08PS steel in 2 M HCl is 105 g·m^{-2}·h^{-1}.

We considered two promising starting products for creating CIs for steel in HCl solutions: Catamin AB (49–51% aqueous solution of a mixture of alkylbenzyldimethylammonium chlorides $[H_3C–(CH_2)_n–N^+(CH_3)_2CH_2C_6H_5]Cl^-$, where $n = 9–17$) and cocamidopropyl betaine (46–48% aqueous solution of lauramidopropyl betaine $H_3C–(CH_2)_{10}–CO–NH–(CH_2)_3–N^+(CH_3)_2–CH_2–COO^-$) (Table 1). The first surfactant is cationic, while the second one is amphoteric. Catamin AB [21–24] and compounds with similar structures [25–28] were previously studied as CIs of steel in solutions of hydrochloric and sulfuric acids and in mineralized aqueous media. Cocamidopropyl betaine (CAPB) is obtained from coconut oil, a natural raw material [29, 30], and used to obtain detergents [31, 32]. Data on the effect of this surfactant on the human organism and on the environment are available [33–35]. CAPB has been less studied as a CI than Catamin AB. It was reported [36] to be less efficient in the protection of carbon steel in stratum water than cocamide diethanol amine and cetrimonium chloride.

Compounds available to us and belonging to anionic and non-ionogenic surfactants were studied as comparison objects (Table 1). The first group is represented by lauroyl sarcosinate ($[H_3C–(CH_2)_{10}–CO–N(CH_3)–CH_2–COO^-]Na^+$, SLS), sodium oleoyl sarcosinate ($[H_3C–(CH_2)_7–HC=CH–(CH_2)_7–CO–N(CH_3)–CH_2–COO^-]Na^+$, SOS) and sodium dodecylbenzenesulfonate ($[H_3C–(CH_2)_{11}–C_6H_4–SO_3^-]Na^+$, SDBS). SDBS is known as a corrosion inhibitor of mild steels in acid sulfate media [37]. The second group of surfactants is represented by the indirect material OP-10:



Combinations of OP-10 with nitrogen-containing compounds are contained in mixed CIs of steels in HCl solutions [38]. Moreover, a nitrogen compound, urotropine ($C_6H_{12}N_4$), has been studied as an additive that can improve the protective effect of surfactants on the corrosion of steels. Urotropine is known as a CI of steels in acid solutions, a component of mixed CIs, and a raw material for the chemical synthesis of CIs, including those manufactured on industrial scale [38–47].

In this study, it is necessary not only to consider the feasibility of steel protection in HCl solutions with individual surfactants but also to analyze whether mixed CIs can be used. This is a promising approach because it often allows one to reduce the consumption of the most expensive components in a mixed CIs without decreasing the protective effect significantly. In some cases, the protective effect of a mixed CI may be higher than that of its most efficient individual component.

Experimental

The corrosion rate was measured for 08PS low-carbon steel (composition, mass %: C –0.08; Mn – 0.5; Si – 0.11; P – 0.035; S – 0.04; Cr – 0.1; Ni – 0.25; Cu – 0.25; As – 0.08). It was determined from the weight loss of strap-shaped samples 50 mm×20 mm in size (at least 3 samples per point) in the media being studied, using 50 mL of the acid solution per sample. The corrosion rate was calculated from the mass loss of the metal samples:

$$k = (m_0 - m_\tau)\cdot S^{-1}\cdot \tau^{-1}$$

where m_0 is the mass of a metal sample before the corrosion test; m_τ is its mass after the test; S is its surface area; and τ is the duration of the corrosion test. The duration of the experiments was 2 hours. Before an experiment, the samples were cleaned on an abrasive wheel (ISO 9001, grit 60) and degreased with acetone.

The efficiency of the CIs was estimated by the corrosion inhibition factors $\gamma = k_0 (k_{\text{in}})^{-1}$ and degree of protection $Z = (k_0 - k_{\text{in}}) \cdot k_0^{-1} \cdot 100\%$, where k_0 is the corrosion rate in the background solution and k_{in} is the corrosion rate in a solution with an agent in question. To estimate the effect of an agent on a surfactant, we calculated the coefficient of mutual effect of the mixture components [2]:

$$K_{\text{m}} = \gamma_{\text{mix}} (\Pi\gamma_{\text{i}})^{-1}$$

where γ_{mix} is the corrosion inhibition factor for a mixture of CIs and $\Pi\gamma_{\text{i}}$ is the product of the corrosion inhibition factors γ_{i} for the individual mixture components. For $K_{\text{m}} < 1$, the mutual protective effect of the components is reduced; for $K_{\text{m}} = 1$, it is additive; and only at $K_{\text{m}} > 1$ does mutual enhancement of protection take place.

Solutions were prepared using HCl (reagent grade) and distilled water. The basic concentration of the surfactants was 5 mM. The concentrations of surfactants in HCl solutions are given in terms of the active substance.

Results and Discussion

The corrosion of 08PS steel in 2 M HCl (60°C) occurs at a high rate, namely 105 $\text{g} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$. Under these conditions, only three of the products studied (Catamin AB, CAPB and urotropine) decrease the corrosion rate at least tenfold (Table 1). Of these compounds, only Catamin AB provides a corrosion rate below 5 $\text{g} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$. Anionic and non-ionic surfactants weakly inhibit steel corrosion.

Equimolar mixtures of the compounds studied with cationic and amphoteric surfactants (Catamin AB and CAPB) showed better results than the individual compounds (Table 2). However, antagonism in the action of the components was observed in all the mixtures studied ($K_{\text{m}} < 1$). Other conditions being equal, the mixtures containing Catamin AB provide better steel protection than the mixtures containing CAPB. From our point of view, the Catamin AB + urotropine and CAPB + urotropine formulations are of interest: despite the formal antagonism in the action of the components, they provide the best inhibition of steel corrosion in the corresponding series of mixtures and are interesting for further studies. It should be noted that rather a high protective effect is provided by the Catamin AB + CAPB formulation, but its study is not very promising because relatively cheap urotropine is more suitable for the creation of mixed CIs from the economic point of view.

A study on the effect of the molar ratio of components in the Catamin AB + urotropine and CAPB + urotropine mixtures on the corrosion of 08 PS steel in 2 M HCl (60°C) showed (Figures 1 and 2) that, other conditions being equal, the Catamin AB + urotropine formulations are superior to the CAPB + urotropine mixtures in terms of corrosion inhibition. For both formulations with a 5 mM total content of CIs, the maximum efficiency is observed for mixtures containing 0.1–1 mM Catamin AB or CAPB. At this ratio of components in the mixture, the antagonism in their action is the smallest (Figure 3). For further studies, we chose the following mixtures: 0.5 mM Catamin AB + 4.5 mM urotropine and 0.5 mM CAPB + 4.5 mM urotropine.

Table 2. Effect of surfactant mixtures ($C_{in} = 5$ mM, molar ratio of components 1:1) on the corrosion rate of 08PS steel in 2 M HCl (k), the corrosion inhibition factors (γ) and the coefficient of mutual effect of the inhibitory mixture components (K_m). $t = 60$°C

No.	**Surfactant**	**Cocamidopropyl betaine**			**Catamin AB**		
		k, g·m^{-2}·h^{-1}	**γ***	**K_m**	**k, g·m^{-2}·h^{-1}**	**γ***	**K_m**
1	Cocamidopropyl betaine	–	–	–	3.4	31	0.07
2	Catamin AB	3.4	31	0.07	–	–	–
3	Sodium lauroyl sarcosinate	9.4	11	0.20	2.9	36	0.25
4	Sodium oleoyl sarcosinate	6.7	16	0.20	2.5	42	0.20
5	Sodium dodecylbenzenesulfonate	10	11	0.47	3.0	35	0.57
6	Indirect material OP-10	9.0	12	0.20	2.8	38	0.25
7	Urotropine	4.5	23	0.16	2.4	44	0.12

* The corrosion rate of 08PS steel in 2 M HCl 105 g·m^{-2}·h^{-1}.

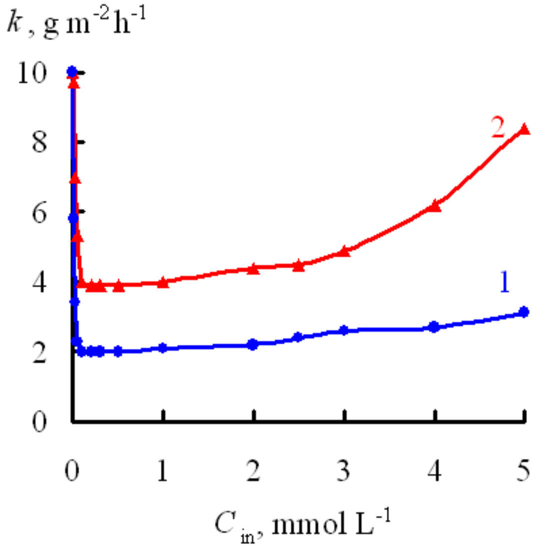


Figure 1. Variation of the corrosion rate of steel 08PS in 2 M HCl containing 5 mM mixtures of Catamin AB + urotropine (1) or CAPB + urotropine (2) *vs*. the concentration of Catamin AB or CAPB. $t = 60$°C.

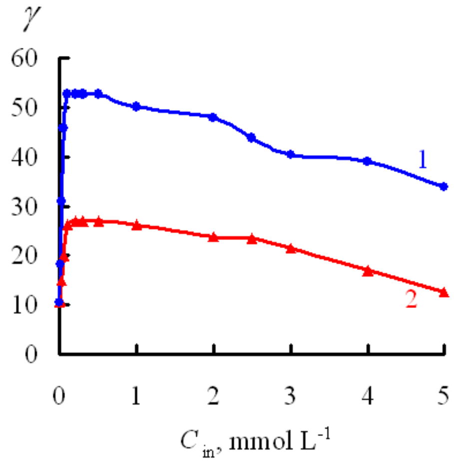


Figure 2. Variation of the corrosion inhibition factors at 08PS steel corrosion in 2 M HCl containing 5 mM mixtures of Catamin AB + urotropine (1) or CAPB + urotropine (2) *vs*. the concentration of Catamin AB or CAPB. $t = 60°C$.

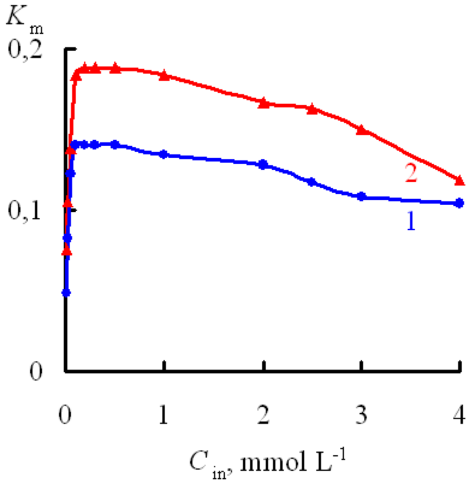


Figure 3. Variation of the coefficient of mutual effect of the inhibitory mixture components on 08PS steel corrosion in 2 M HCl containing 5 mM mixtures of Catamin AB + urotropine (1) or CAPB + urotropine (2) *vs*. the concentration of Catamin AB or CAPB. $t = 60°C$.

In 2 M HCl solution, the plot of the degree of steel protection *versus* the logarithm of the concentration of a CI mixture (Catamin AB + urotropine and CAPB + urotropine, molar ratio of components 1:9) is S-shaped (Figure 4). Judging by this plot, at a content of 0.25 mM of inhibitor formulations in the solution, the maximum coverage of the metal surface with the CIs is observed.

In 2 M HCl, the presence of the composite CIs studied does not fundamentally affect the effective activation energies of corrosion of 08PS steel calculated from the plots of its corrosion rate *versus* temperature (Figure 5). The obtained values of the effective activation energies in all cases indicate that the corrosion process occurs under mixed diffusion-kinetic control. The observed decrease in the effective activation energy of corrosion by the CI mixtures indicate an increase in the fraction of the diffusion component.

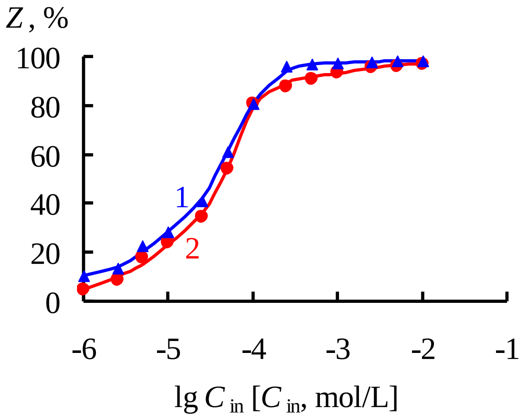


Figure 4. Variation of the degree of protection at 08PS steel corrosion in 2 M HCl containing mixtures of Catamin AB + urotropine (1) or CAPB + urotropine (2) at molar ratio of components 1:9 *vs*. the concentration of mixed inhibitors. t = 60°C.

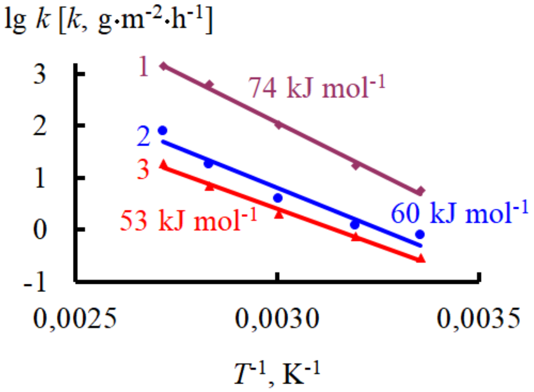


Figure 5. Dependence of lg$k$ on T^{-1} for corrosion of 08PS steel in 2 M HCl (*1*) containing 0.5 mM CAPB + 4.5 mM urotropine (*2*) or 0.5 mM Catamin AB + 4.5 mM urotropine (*3*).

The formulation of 0.5 mM CAPB + 4.5 mM urotropine efficiently inhibits the corrosion of 08PS steel in 2 M HCl at $t \leq 60°C$, providing the maximum $k = 3.9$ g·m^{-2}·h^{-1} (Table 3). At $t = 60°C$, the corrosion of steel slows down by a factor of 27. At higher temperatures, $t = 80$ and 95°C, the corrosion rate of steel decreases 34- and 18-fold, respectively, but the corrosion rates are still high, $k = 18$ and 78 g·m^{-2}·h^{-1}. The formulation of 0.5 mM Catamin AB + 4.5 mM urotropine inhibits steel corrosion more strongly. A significant reduction in the steel corrosion rate in the presence of this formulation is observed at $t \leq 80°C$ where the maximum corrosion rate is $k = 6.9$ g·m^{-2}·h^{-1}. The corrosion process at $t = 80°C$ is inhibited by a factor of 90. At a higher temperature, 95°C, the corrosion of steel is accelerated ($k = 19$ g·m^{-2}·h^{-1}, $\gamma = 74$).

Table 3. Effect of 0.5 mM CAPB + 4.5 mM urotropine and 0.5 mM Catamin AB + 4.5 mM urotropine on the corrosion rate of 08PS steel in 2 M HCl (k, g·m^{-2}·h^{-1}) and the corrosion inhibition factors (γ).

Inhibitor	**Factor**	**Temperature, °C**				
		25	**40**	**60**	**80**	**95**
–	k	5.7	17	105	620	1400
CAPB + urotropine	k	0.77	1.2	3.9	18	78
	γ	7.4	14	27	34	18
Catamin AB + urotropine	k	0.29	0.74	2	6.9	19
	γ	20	23	53	90	74

The possibility of improving the protective effect of the formulations at $t = 80$ and 95°C should be analyzed. It has been found that an increase in the relative content of surfactants in the mixtures studied worsens the protection of steel. For example, the mixtures of 2 mM CAPB + 3 mM urotropine and 2 mM Catamin AB + 3 mM urotropine give values of $k = 21$ and 10 g·m^{-2}·h^{-1}, respectively, which are smaller than those of the formulations with the 1:9 ratio of the components. The use of formulations containing 10 mM of a mixed inhibitor did not improve the protective effect on steel significantly (Table 4). For both composite inhibitors, the most optimal surfactant/urotropine ratio of the components is 1:9. Attempts to increase the content of the mixed inhibitor in the corrosion medium significantly are also unpromising. In the presence of 5 mM CAPB + 25 mM urotropine and 5 mM Catamin AB + 25 mM urotropine, the values of k are 11 and 7.0 g·m^{-2}·h^{-1}, respectively. It should be noted that the formulations containing 1 mM CAPB + 9 mM urotropine and 1 mM Catamin AB + 9 mM urotropine that are most efficient at $t = 80$°C provide $k = 50$ and 18 g·m^{-2}·h^{-1}, respectively, at $t = 95$°C. This result is not fundamentally different from the corrosion rates observed for formulations of 0.5 mM surfactant + 4.5 mM urotropine at the same temperature. Thus, the optimal formulations for the protection of steel in HCl solutions are mixtures of Catamin AB or CAPB with urotropine at the 1:9 molar ratio of the components. The total content of the composite CI in the corrosive medium should be 5 mM. Increasing the total content of the composite CI in the corrosive medium or the fraction of the most expensive components (Catamin AB or CAPB) leads to an unjustified increase in the inhibitor consumption without any significant gain in metal protection.

08PS steel in HCl solutions at temperatures up to 60°C, inclusively, can be protected by formulations of 0.5 mM CAPB + 4.5 mM urotropine. It is important that in industrial operation of this acid solution, its temporary overheating to $t = 80$°C will not lead to a significant decrease in the protective effect of the mixed inhibitor. More interesting is the composite CI comprising 0.5 mM Catamin AB + 4.5 mM urotropine, which efficiently slows down steel corrosion at temperatures up to 80°C, inclusively. This inhibitor formulation is resistant to a temporary overheating of the HCl solution up to $t = 95°C$ while maintaining a satisfactory inhibitive effect.

Table 4. Effect of inhibitor mixtures ($C_{in} = 10$ mM) on the corrosion rate of 08PS steel in 2 M HCl (k, $g \cdot m^{-2} \cdot h^{-1}$) and the corrosion inhibition factors (γ). $t = 80°C$.

Surfactant	Factor	Surfactant concentration, mM					
		10	**0.5**	**1**	**2**	**4**	**0**
Cocamidopropyl betaine	k	44	13	12	12	14	150
	γ	14	48	52	52	44	4.1
Catamin AB	k	21	6.6	6.6	6.9	8.1	150
	γ	30	94	94	90	77	4.1

* The corrosion rate of 08PS steel in 2 M HCl is 620 $g \cdot m^{-2} \cdot h^{-1}$.

The results obtained in this study are important from the theoretical and practical points of view. Composite two-component CIs for the protection of low-carbon steel in HCl solutions have been developed. The uniqueness of these two-component CIs is determined by the fact that their maximum protective effect is manifested at a very low content of the component which is the strongest corrosion inhibitor in individual state. At the same time, the composite CI significantly slows down steel deterioration more strongly than any of the individual mixture components at an equivalent content in the corrosive medium. The observed effect is interesting and requires further additional studies using the capabilities of various physicochemical and electrochemical research methods.

Conclusions

1. The protective effect of the surfactants we studied on the corrosion of 08PS steel in HCl solutions decreases in the series: Catamin AB > CAPB > SOS > OP-10 > SLS > SDBS. Of these surfactants, cationic and amphoteric surfactants provide the maximum corrosion inhibition.
2. The most efficient way to use surfactants for steel protection in acidic media is to create mixed CIs on their basis. This approach makes it possible to increase the efficiency of steel protection and reduce the consumption of CI components.
3. The formulations of 0.5 mM CAPB + 4.5 mM urotropine and 0.5 mM Catamin AB + 4.5 mM urotropine are recommended for the protection of mild steel in HCl solutions with temperatures up to 60 and 80°C, respectively.

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