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THERMAL TREATMENT OF CONCENTRATED LIQUID TOXIC WASTE AND AUTOMATIC CONTROL OF PROCESS EFFICIENCY

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Розроблена енерготехнологічна система і спосіб термічної переробки рідких токсичних відходів на основі глибокого термічного упарювання в апаратах заглибного горіння. Встановлено, що щільність і в'язкість РТВ можна використовувати для оцінки ефективності процесу термічного упарювання. Розроблено метод контролю щільності і в'язкості РТВ, що упарюються вібраційним методом з використанням занурювального механічного резонатора. Наведено блок схему розрахунку щільності і в'язкості при багатоконтурному контролі параметрів

Ключові слова: рідкі токсичні відходи, термічне упарювання, аппарат зануреного горіння, вібраційний метод контролю, механический резонатор

Разработана энерготехнологическая система и способ термической переработки жидких токсичных отходов на основе глубокого термического упаривания в аппаратах погружного горения. Установлено, что плотность и вязкость ЖТО можно использовать для оценки эффективности процесса термического упаривания. Разработан метод контроля плотности и вязкости упариваемых ЖТО вибрационным методом с использованием погружного механического резонатора. Приведена блок схема расчета плотности и вязкости при многоконтурном контроле параметров

Ключевые слова: жидкие токсичные отходы, термическое упаривание, аппарат погружного горения, вибрационный метод контроля, механический резонатор

1. Introduction

Liquid toxic waste (LTW) accounts for 30 to 50 % of the total amount of wastewater entering the basins of Ukraine [1]. Development of nuclear power and extension of the scope of radioactive isotopes in various fields of science and technology are associated with pollution of natural waters with radioactive waste. Radioactively contaminated waters are characterized by a variety of radioactive elements contained in them. In addition, up to 1 km^3 of liquid toxic wastewater containing 50 thousand tons of heavy metals, 100 thousand tons of acids and alkalis (waste of etching and electroplating departments and production units of various industries) is annually released into the environment [2].

Minimization of the volume and activity of waste due to improvement of energy-efficient treatment technologies is one of the urgent tasks in environmental protection.

2. Literature review and problem statement

Treatment of liquid toxic waste is aimed at solving two main problems: cleaning of the bulk of waste of radionuclides, heavy metal salts and concentration of the latter in the minimum volume [3]. To this end, three groups of cleaning methods are used: thermal, sorption, membrane [4]. Thermal and sorption methods are well developed and widely used in practice. Currently, they are the basis of operation of treatment facilities for processing waste from various nuclear reactors, wastewater of etching and electroplating departments [5].

Thermal methods involve the use of heat to concentrate waste by transforming the main component – water – into steam. The main thermal methods are thermal distillation (evaporation) and drying [3]. Drying is commonly used for preparation (dehydration) of LTW concentrates for solidification. Thermal distillation differs in the nature of vaporization (volume boiling or surface evaporation). As a heat carrier, steam, hot gases, electricity, organic products can be used. Heat supply can be carried out by direct contact with the heat carrier or heat transfer through the device wall. In the practice of waste treatment, vaporization distillation when boiling with the heat supply by water vapor through the evaporator wall is most widely used. Such process ensures heat transfer in the absence of contact of the clean heat carrier with the radioactive process liquor.

Unlike other methods (sorption and membrane), thermal distillation allows condensate cleaning of radionuclides in any form: ionic, molecular or colloidal [6, 7]. In this case, cleaning is limited only by the volatility of radionuclides. Low requirements to the quality of the waste subject to thermal distillation (presence of colloids, detergents) allow excluding the use of special sedimentation operations before it. This sets thermal distillation apart from sorption (dynamic) and some membrane methods. And, finally, the possibility of obtaining high cleaning factors allows thermal distillation, if necessary, to solve the problem of condensate treatment to waste standards or circulating water norms independently and completely [8]. Distillation allows achieving a high degree of cleaning of radionuclides ($Kcl=1\cdot10^{-6}$).

The possibility of obtaining high cleaning factors makes the tasks of developing new thermal distillation technologies very relevant. The majority of wastewater thermal distillation technologies known today are associated with the use of the evaporation process with mechanical vapor recompression [9]. The developed evaporation systems based on such technologies are very power-intensive and expensive [10].

One of the promising approaches to reducing the energy consumption of LTW evaporation is the use of modern energy-efficient thermal devices. Among them, a special place is occupied by immersion combustion units (ICU). The devices and plants operating on their basis are characterized by the efficiency of more than 100 % relative to the lower calorific value [11].

Literature review has shown that the method of LTW thermal distillation (evaporation) in volume by means of immersion combustion units is not applied in the industry [12]. The paper [12] describes the results of the immersion evaporator development and laboratory tests that allow the unit to be used in the thermal distillation technology.

Due to the lack of thermal distillation method with the use of ICU, no data on universal methods for monitoring the LTW distillation process efficiency have been found in the literature. A number of works touch upon the issue of monitoring individual parameters of the wastewater treatment process: concentration and rate of evaporation [13], temperature and pressure in the unit [14]. The vibration method for controlling the oscillations of the gas-liquid system in the immersion combustion unit has been described in [15], but the possibility of using the method for monitoring the pulsations of liquid radioactive wastewater has not been investigated.

Thus, the works and research related to the development of an LTW treatment method using the immersion combustion unit and process efficiency control methods should be recognized as a promising aspect of solving environmental safety problems and introducing modern energy-efficient technologies in Ukraine.

3. The aim and objectives of the study

The aim of the paper is to develop an energy-efficient technological system (EETS) that would ensure waste cleaning of radionuclides and heavy metal salts with a high degree of waste concentration, with the continuous control of process efficiency.

To achieve the aim, the following objectives were accomplished:

– to develop the energy-efficient LTW treatment method, allowing to increase the degree of waste concentration, to select the equipment that prevents salt encrustation on the evaporator and plant heating surfaces;

– to transform the salt concentrate into a fire-explosion-proof condition and to simplify the LTW treatment procedure in general;

– to develop a scheme for automatic control of the LTW distillation process efficiency;

– to develop a design of the turbulent pulsation transducer for monitoring the evaporation density of the medium based on the vibration method.

4. Description of the liquid toxic waste thermal treatment plant

The basis for the design was the liquid radioactive waste (LRW) thermal treatment plant based on the ICU. The plant was developed for the experimental nuclear reactor of the Institute for Nuclear Research of the Academy of Sciences of Ukraine (Ukraine), tested and commissioned [15].

To solve the set-out problem, it was proposed to perform thermal distillation in the ICU to the level of deep evaporation and drying in a specially designed radiation chamber.

The developed EETS for the thermal treatment of LTW and wastewater of etching and electroplating production units (Fig. 1) is a heat-insulated refractory fireproof chamber (1) in the form of a ∅1800 mm vertical stainless steel cylinder with the ICU located in it. The chamber is equipped with an explosion relief valve (2), mounted in the upper part of its casing. In addition, the EETS is equipped with an autonomous (capacity of 1580 m^3/h) blower, LTW storage tank (3), pump unit (4), EC&I unit. In the central part along the axis of the fire chamber, there are a flat-flame burner GPPs-5 (fuel consumption of 160 m^3/h, pressure of 12 kN/m^2) with the fuel and combustion air supply system and shut-off equipment. Gas piping and shut-off equipment are complete with measuring sensors.

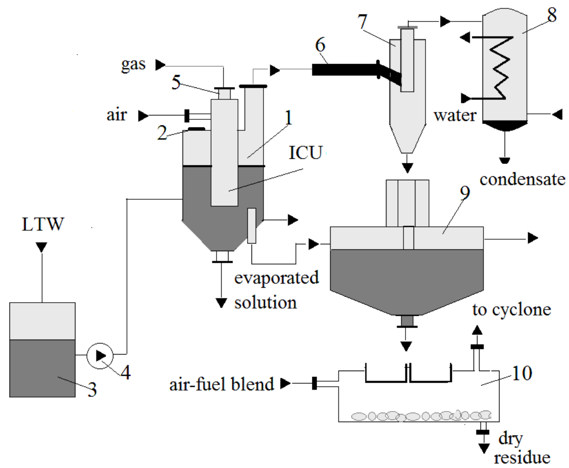

Fig. 1. LTW thermal treatment plant based on the ICU: 1 – heat-insulated fireproof chamber; 2 – explosion relief valve; 3 – LRW storage tank; 4 – pump unit; 5 – burner unit; 6 – gas duct; 7 – cyclone; 8 – condenser; 9 – sedimentation tank; 10 – radiation chamber

The control system (EC&I cabinets) is designed for automatic LTW feeding into the fire chamber, radioactivity monitoring, signaling and protection of equipment.

Combustion products are removed from the fire chamber through the gas duct (6), cyclone (7) with a capacity of 5600 m^3/h, condenser (8), smoke exhauster with a capacity of 6,800 m^3/h, connected to a chimney.

The technology of gaseous waste treatment depends on the type of waste. In some cases, gaseous waste is deactivated prior to be released into the atmosphere by passing through electric dust precipitators or filters made of glass wool, cloth or asbestos. For the release of gaseous radioactive substances, the emissions are passed through chemical absorbers. If there are short-lived isotopes in the emissions, they are sometimes held in gas holders (large cylinders) until the complete disintegration. The emissions, deactivated in one way or another, containing only traces of radioactive substances are removed through high pipes for better dispersion in the atmosphere.

In addition, the EETS is equipped with a sedimentation tank (9) with a stirrer, electric motor and radiation chamber (10) to dry the evaporated LTW in the fire chamber to a dry residue state.

The entire set of the EETS equipment is located in a separate building, with the LTW feeding pipeline system connected to the LTW storage tank with a room for the operator. The gas piping is assembled in accordance with the requirements, control and regulation of pressure values at the burner inlet are provided.

The plant operates as follows. The fire chamber of LTW is filled to the specified level. The ICU burner plug is ignited and steady combustion is achieved. As a result of fuel combustion, flue gases containing N_2, O_2, C are formed, as well as molecules of harmful substances, the atmospheric emission of which is not allowed.

Flue gas cleaning takes place in the cyclone with the deposition of harmful substances into the hopper and then purified flue gases go to the chimney through the condenser and gas duct.

The developed EETS allows effective treatment of LTW with a volume of ~1,000 l/h [15]. During the evaporation in the ICU, the end products remained liquid at ρ=1.3–2.0 g/cm^3 and the salt content of 1,300–2.000 g/l of the product, which is almost 2 times higher than evaporation indices of the devices used in the practice of salt LRW processing (650 g/l). In this case, evaporation of salt crystals in the fire chamber begins when the salt content reaches 800–1.000 g/l [15].

The positive fact is that the proposed LRW processing method allows a stage of deep concentration to be performed practically without salt encrustation on heat transfer surfaces, which is due to design features of the ICU combustion chamber [15].

From the analysis of experimental data (Table 1), it follows that the concentration of LTW components (heavy metal ions) increased on average 2.5–3 times as a result of the thermal treatment process. The chemical composition of LTW, subjected to thermal treatment, and dry residue (after the radiation chamber) has not changed.

Energy efficiency was estimated based on the operation of the immersion combustion unit, the efficiency of which is 0.98 [16].

Table 1

Chemical composition and concentration of substances in the stages of solution and dry residue

Experimental stage	Concentration, g/l										
	pH	Mg^{2+}	Cu^{2+}	Pb^{2+}	Fe^{2+}	Cr	Zn^{2+}	Ni^{2+}	Ca^{2+}	Co^{2+}	Al^{2+}
Solution of clarifying red-copper pipes											
I	1.35	0.19	35.9	0.17	0.026	6.32	82	0.94	1.13	0.4	0.09
Solution after neutralization											
II	11.3	58.5	4.1	7.3	0.22	156.3	6.7	43.3	3.6	125	7.5
Solution after treatment											
III	–	0.19	–	0.14	0.03	8.4	15	0.23	0.39	0.15	1.0
Evaporated sludge											
I	2.0	0.22	34.4	0.2	0.026	8.95	82	1.2	0.45	0.013	–
II	9.9	53.2	31.7	7.6	0.28	153.8	19.3	4.3	108	–	6.3
III	–	0.32	–	0.39	0.03	21.9	18.4	0.46	0.15	0.15	27.3
Dry sludge (5 % solution)											
I	2.6	0.67	103	0.6	0.04	33.6	20.8	3.4	0.7	0.013	–
II	7.0	72.6	43.5	7.6	0.03	131	73.3	3.9	80.4	–	6.3
III	–	0.61	–	0.91	0.06	75.8	45.6	1.4	0.58	0.15	–

5. Vibration method of density and viscosity control of evaporated LTW

One of the indicators of efficiency of the LTW distillation unit are the density and viscosity of evaporated effluents. Control of the concentrated LTW treatment process is impossible without the application of a reliable method for monitoring these parameters. Density characterizes the quality of an output product and is subject to mandatory control in the automatic control system of feeding the evaporated LTW solution in the radiation chamber after treatment in the ICU. The value of the density of evaporated effluents is the main efficiency criterion of the LTW drying and crystallization processes.

Control of density and viscosity of the evaporated solution is made by the vibration method. The problem of simultaneous measurement of the LTW density and viscosity by vibration methods is a significant (up to 50 %) decrease in the quality factor of the mechanical resonator due to the damping of its vibrations by a viscous medium.

The operation principle of the mechanical resonator of the "vibration dumbbell" type (Fig. 2) consists in the following: when the controlled oscillator 9 is switched on in the windings 7, 8, a current pulse appears, which causes opposing longitudinal magnetic fluxes F_{OP1} and F_{OP2} in the magnetoelastic rods 2, 3. At the same time, the rotating magnetic fluxes F_{W1}, F_{W2}, caused by a direct current in the winding 6, act on the rods 2, 3. The interaction of the magnetic fluxes causes the appearance of counter-oscillations of the rods 2, 3, and of the cylinders 4, 5 together with them. In this case, in the nonmagnetic connector there is a nodal line, which remains fixed throughout the vibration process. In the nodal line, the resonator is fixed to the column 11. The moments of the forces acting on the cylinders 4, 5 linearly depend on the magnitude of direct current I_c of the winding 6. The resonator vibration amplitude changes with the current in the winding 6. The vibration frequency of the oscillator 9 can be controlled by the resonator vibrations. To convert the resonator vibrations into an electrical signal, the receiver 10 is used.

The intermediate measuring transducer (Fig. 3) contains a vibration frequency control circuit and a resonator damping control circuit. The resonator transmission function can be expressed as:

$$W_p(p,t)=\frac{k}{m(t)\cdot p^2+h(t)\cdot p+c}, \tag{1}$$

where $m(t)=m_0+m_{ad}(t)$ is the total mass of the movable part of the resonator; m_0 is the resonator mass; $m_{ad}(t)$ is the added mass of the liquid; $h(t)=h_0+h_{ad}(t)$ is the resonator vibration damping factor; h_0 is the damping of the free resonator from energy losses due to internal friction in the material and in the place of fixing; $h_{ad}(t)$ is the added damping (damping by a liquid); c is the resonator rigidity, k is the static transmission factor of the resonator.

For cylinders with radial partitions (Fig. 3), with small amplitude vibrations, the dependence of the added mass $m_{ad}(t)$ and the damping factor $h_{ad}(t)$ on the measured density ρ, viscosity η and vibration frequency $\omega'=\omega_0+\Delta\omega$ has the form:

$$\left.\begin{aligned} m_{ad}&=\alpha_1+\alpha_2\cdot\sqrt{\rho\cdot\eta\cdot\omega'},\\ h_{ad}&=\alpha_3\cdot\sqrt{\rho\cdot\eta\cdot\omega'},\end{aligned}\right\} \tag{2}$$

where α_1, α_2, α_3 are constant coefficients (determined experimentally), ω_0 is the initial circular vibration frequency of the resonator, $\Delta\omega$ is the change in the resonator vibration frequency.

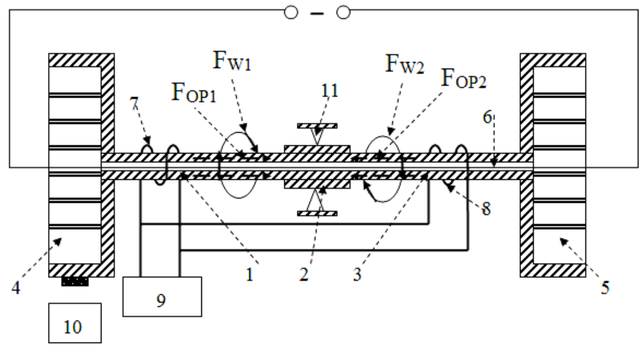

Fig. 2. Resonator of the immersed measuring transducer of liquid density and viscosity: 1 — elastic non-magnetic connector; 2, 3 — magnetoelastic rods; 4, 5 — cylinders with partitions; 6 — winding of the DC voltage passing through an internal cavity of both rods; 7, 8 — coaxial windings; 9 — controlled generator; 10 — receiver of torsional vibrations; 11 — resonator fixing column

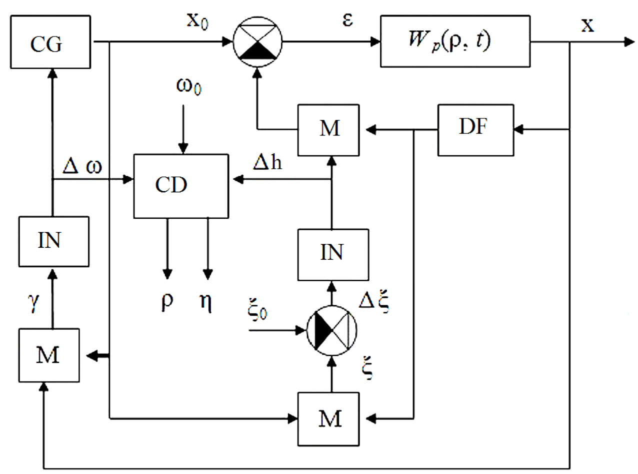

Fig. 3. Block diagram of the intermediate measuring transducer: CG — controlled generator; IN — integrator; M — multiplier; DF — differentiation unit; CD — computing device; x, ε — system coordinates; ρ, η — controlled density and viscosity; *p* —differentiation operator; $x_0=\alpha_0\sin(\omega_0+\Delta\omega)$ — input signal; γ, ξ — control coordinates of the adaptation circuits; $W_p(\rho, t)$ — resonator transmission function; *t* — time coordinate

The dynamics of operation of the intermediate measuring transducer (Fig. 3) can be represented by the following system of differential equations:

$$\left.\begin{aligned} &\left[m(t)\cdot p^2+(h(t)+\Delta h)\cdot p+c\right]\cdot x=a_0\cdot\sin\omega'\cdot t,\\ &\Delta\omega=k_u\cdot\frac{x_0\cdot x}{p},\ \Delta h=k_u\cdot\frac{x_0\cdot\dot{x}-\xi_0}{p},\end{aligned}\right\} \tag{3}$$

where k_u is the integrator coefficient, x is the angular velocity of the resonator, a_0 is the vibration excitation force amplitude.

The parameters $m(t)$, $h(t)$ are quasistationary, namely, changes in the vibration frequency Δω and parametric compensation coefficient *Δh* during the transient process are small, which can be ensured by selecting the integrator coefficient k_u. The solution of the system of equations has the form:

$$x = A\cdot\sin(\omega'\cdot t+\phi), \tag{4}$$

where the values of A and ϕ can be found from the expressions:

$$A=\frac{a_0\cdot k}{\sqrt{(c-m\cdot\omega'^2)^2+(h+\Delta h)^2\cdot\omega'^2}},$$

$$\phi = arctg\frac{(h+\Delta h)\cdot\omega'}{c-m\cdot\omega'^2}.$$

The control coordinates of the adaptation circuits $\gamma=x_0\cdot x$, $\xi=x_0\cdot x$ with allowance for (3) can be represented as:

$$\left.\begin{aligned}\gamma &= k\cdot\frac{a_0^2}{2}\cdot\frac{c-m\cdot\omega'^2}{(c-m\cdot\omega'^2)^2+(h+\Delta h)^2\cdot\omega'^2},\\ \xi &= -k\cdot\frac{a_0^2}{2}\cdot\frac{(h+\Delta h)\cdot\omega'^2}{(c-m\cdot\omega'^2)^2+(h+\Delta h)^2\cdot\omega'^2}.\end{aligned}\right\} \tag{5}$$

The intermediate measuring transducer supports the system in the resonance mode by means of the CG (Fig. 3). To determine the nature of the dependences (5), we take $c-m\cdot\omega'^2=\lambda$, where λ is a rather small value proportional to a frequency deviation from the resonance frequency. In view of the above, the relationship can be represented as:

$$\left.\begin{aligned}\gamma &= k\cdot\frac{a_0^2}{2}\cdot\frac{\lambda}{(h+\Delta h)^2\cdot\omega'^2},\\ \xi &= -k\cdot\frac{a_0^2}{2}\cdot\frac{1}{(h+\Delta h)},\end{aligned}\right\} \tag{6}$$

where Δh is the parametric compensation of the resonator damping.

In the initial setting, the coordinate of the system is $\xi_0=-0.5\cdot k\cdot a_0^2$. Then the relation for $\Delta\xi=\xi-\xi_0$ can be represented as:

$$\Delta\xi=\xi_0\cdot\frac{h_{np}+\Delta h}{h+\Delta h}. \tag{7}$$

Around the resonance, the coordinate γ is proportional to a resonance frequency deviation from the CG frequency, and the coordinate $\Delta\xi$ is proportional to the sum of the added damping h_{ad} and parametric compensation Δh.

In the presence of information on h_{ad}, the exact values of density ρ and viscosity η of the controlled LTW are calculated (Fig. 4).

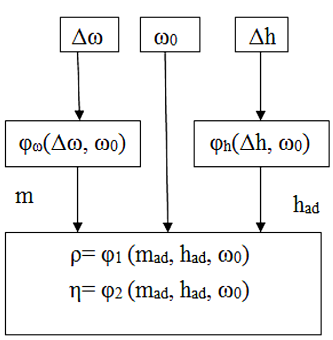

Fig. 4. Block diagram for density and viscosity calculation in multi-circuit control of parameters

The selected algorithms for forming the coordinates γ, ξ and the presence of integrators in the adaptation circuits provide continuous resonance setup of the system while maintaining the resonator quality factor. This allows considering the output signals of the integrators $\Delta\omega$, Δh definitely related to the added mass m_{ad} and the added damping factor h_{ad}. The computing device processes signals of the parametric circuits.

6. Discussion of the results of the study of the energy-efficient technological system of LTW treatment

The developed EETS and the method of LTW thermal treatment on the basis of deep thermal evaporation in the ICU in comparison with the existing thermal treatment technologies [10] have significant advantages:

– almost 2–3 times (from 600 g/l to 1.300–2.000 g/l) higher degree of concentration of radioactive waste;

– prevent salt encrustation on the heating surfaces of the ICU and the plant as a whole;

– simplify the vat residue treatment operation (preliminary technological and mechanical preparation before thermal treatment in the ICU is not required);

– allow obtaining the final product in the form of a dry residue, which eliminates the embedment (hardening) of radioactive salt concentrates to the technological product, preventing the release of radionuclides into the environment;

– obtaining the final product in the form of dry radionuclide salts, which can be returned back to the technology, or subject to disposal in special tanks.

It is determined that the density and viscosity of evaporated effluents can be used as efficiency indicators of the LTW distillation complex. The proposed vibration control method allows calculating simultaneously the density and viscosity values of controlled LTW. The selected algorithms for forming the coordinates of the presence of integrators in the adaptation circuits provide continuous resonance setup of the system while maintaining the resonator quality factor.

7. Conclusions

1. The energy-efficient method for LTW treatment is developed, in which thermal distillation is carried out in the ICU to a degree of deep evaporation and drying in a specially designed radiation chamber. The design of the ICU radiation chamber allows the stage of deep concentration to be made practically without salt encrustation on heat transfer surfaces.

2. Obtaining the final product in the form of a dry residue does not require additional drying and holding operations before disposal or landfilling. The proposed flow diagram of thermal distillation does not require also preliminary technological and mechanical preparation of the vat residue before thermal treatment in the ICU.

3. It is found that the density of evaporated effluents is the main efficiency criterion of the LTW drying and crystallization processes. The method for nondestructive control of toxic waste density using the multi-circuit scheme for auto-

matic monitoring of amplitude-frequency characteristics of the medium is proposed.

4. For the control of turbulent pulsations in a liquid toxic medium, the design of an immersion sensing element of the vibration-frequency sensor and intermediate transducer is proposed. The structure of the automatic density control scheme allows compensating for a decrease in the resonator quality factor due to the damping of its vibrations by a viscous medium.

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