# CONCENTRATION OF EXTRACTION LIQUIDS OF TRADITIONAL CHINESE MEDICINES IN AN INDUSTRIAL SCALE FLUIDIZED BED EVAPORATOR

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## ABSTRACT

In order to inhibit foulant deposition on the inner surface of steam heating tubes in an evaporating concentrator of traditional Chinese medicines, solid particles were introduced into the tubes and a vapor-liquid-solid fluidized bed evaporator was developed. The results of fouling inhibition and operational feasibility have been confirmed in a lab-scale and a pilot size. In this work, an industrial scale fluidized bed evaporator with heat transfer area of heater of 18.9 $m^2$ was designed and established based on the previous investigations. It was applied in the concentration of extracts of several typical traditional Chinese medicines with high viscosity and density. Zero-fouling experimental results were verified and feasibility of operation was also validated after a long running time with different kinds of extracts. In addition, the applications of such a fluidized bed evaporator to other process industries are also mentioned, in which heat transfer enhancement and fouling prevention are needed.

## INTRODUCTION

In the process industries, such as chemical, petroleum, metallurgy, power, pharmaceutical, food, environment, light industry and so on, there is a troublesome fouling deposition on heat transfer surfaces in heat exchangers. The existence of fouling on heat transfer surfaces reduces the heat transfer efficiency, increases energy and material consumptions, and may bring about safety and product quality risks. According to statistics, cost penalties due to fouling, e.g. for additional fuel, down-time, over-design, cleaning chemicals, etc., have been estimated as about 0.25% of the gross domestic product (GDP) of industrialized nations (Steinhagen, et al., 1990;1993; Muller-Steinhagcn,2000; Malayeri, et al., 2015). Therefore, fouling is an important challenge in the development, design and application of heat exchanger.

Several measures have been put forward to prevent or eliminate fouling in the heat exchangers, including chemical and physical techniques. Chemical methods mainly include adding chemical inhibitor and chemical cleaning. Physical techniques consist of adding solid particles, introducing rubber balls, exerting physical (ultrasonic, optical, electro-or/and magnetic, etc.) fields, disturbing the flow boundary layer of heat transfer tube with rotary device or spring undergoing a reciprocating movement and other inserts, increasing flow velocity, utilizing tube vibration induced by flow of fluid, for evaporator to prevent boiling in the heating chamber by increasing the height of liquid column or reverse circulation, mechanical cleaning, surface modification, applying new heat transfer materials, etc. Among them, the method of adding solid particles into heat exchanger to mitigate or eliminate fouling on heat transfer surface is a pure physical process and is a unique and effective online antifouling and heat transfer enhancing measure. The scale on the heat transfer surface can be prevented online by the scouring effect of fluidized solid particles. For the situation of no scale being deposited on the heat transfer surface, adding solid particles can enhance heat transfer process due to thinning of flow and thermal boundaries resulted from the collisions between heat transfer walls and the fluidized solid particles. The technology of fluidized bed heat exchange can be applied to many types of heat exchangers, such as evaporator, boiler, reboiler, crystallizer, condenser, and preheater and cooler, etc., especially to the evaporator that often undergoes a serious fouling problem due to solution boiling and evaporation.

Fluidized bed heat exchangers, first investigated in the USA in 1960s as brine heaters in seawater desalination, have been developed during the last 50 years in the USA, Netherlands, Germany, Japan, South Korea and China, et al (Aghajani, et al., 2005; Hatch, et al.,1966; Iida, et al.,2002; Klaren, 1975; Klaren and De Boer, 2011; Lee, et al.,2003; Liu, et al.,2004; Rautenbach, et al.,1991; Zhang and Li, 2000). Among the researchers, Klaren and his research group have continuously focused on the development and marketing of the fluidized bed heat exchanger and have made a big progress in designing or revamping existing heaters, evaporators, reboilers and crystallizers into a self-cleaning configuration since 1975 (Klaren, 1975; Klaren and De Boer, 2011). We have also continuously explored a fluidized bed evaporator since 1980s (Liu, et al., 2004; Zhang and Li, 2000).

It is known that the fluidized bed heat exchanger is a relatively broad concept, and there are several structural styles, operation modes and application media. The fluidized bed evaporators developed in our research group are mainly the installations of natural circulation type or vertical long-tube natural circulation evaporators and this type of the evaporator hosts the features of low power consumption, high operation steadiness and simple structure. We developed it to solve the fouling problems existing in the process industries.

In the past years, we had developed lab-scale (with heat transfer area of 0.3 $m^2$) and pilot scale (with heat transfer area of 1.7 $m^2$) fluidized bed evaporators with the type of natural circulation flow to prevent the fouling of heat transfer surface in the evaporation process of high viscosity and density extraction liquids of traditional Chinese medicines, and good evaporation results had been achieved (Liu, et al., 2004; Liu, et al., 2005).

Recently, a fluidized bed evaporator with industrial scale (with heat transfer area of the heater of 18.9 $m^2$) has been built and the experiments of concentration of extraction liquids of traditional Chinese medicines with high viscosity and high density have been carried out. The aim of the investigations is to overcome the deficiency of the traditional Chinese medicine evaporation concentration technology, and to provide a technique, an installation and a method for solving the surface fouling phenomenon in the process of the evaporating concentration of the traditional Chinese medicine extracts. The experimental results of concentration of Gengnian'an and other traditional Chinese medicine extraction liquids with high viscosity and high density in the industrial scale vapor-liquid-solid fluidized bed evaporator will be reported in this paper. Other new examples of industrial applications of the fluidized bed evaporator will also be briefly shown. At last, some fundamental understandings on the fluidized bed evaporators will be discussed.

## EXPERIMENTAL

An industrial operation unit of fluidized bed evaporation with a natural circulation vapor-liquid-solid boiling flow was designed and established to concentrate the extraction liquids of some traditional Chinese medicines with high viscosity and density continuously based on the previous investigations on the characteristics of vapor-liquid-solid flow, and heat transfer and fouling prevention in the lab-scale and pilot-scale fluidized bed evaporation installations. The schematic diagram of the fluidized bed evaporation unit system is shown in Fig. 1 and the system is similar to the pilot scale one (Liu, et al., 2005).

When the extraction liquid in the industrial scale fluidized bed evaporation system is heated by the boiler steam to the temperature of boiling point of solution, a vertical vapor-liquid-solid natural circulation flow will be formed. In this condition, continuous collisions between solid particles fluidized by vapor-liquid flow in the heating tubes and the inner wall surface of the heating pipe will exist, thus break the flow and heat transfer boundary layers on the wall surface and inhibit the deposition of fouling on the inner walls of the heating tube. The evaporation system

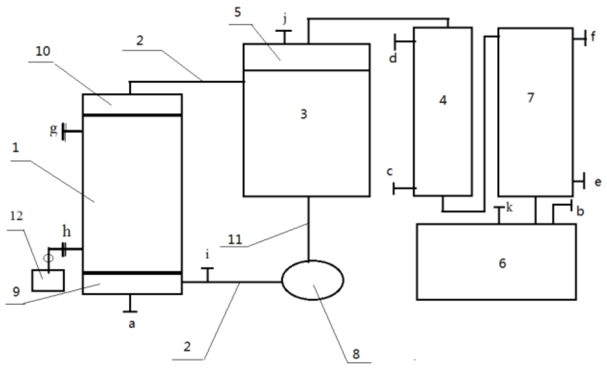


Fig.1. Experimental setup of industrial scale fluidized bed evaporator for extraction liquids concentration of traditional Chinese medicines with a vapor-liquid-solid boiling flow. 1-Shell-and-tube fluidized bed heater；2-connecting pipe；3-vapor-liquid separator；4- plate type condenser of vapor；5-wire mesh demister；6-condensate gauge bank of vapor；7- shell-and-tube type condenser of vapor；8-discharge outlet；9-lower tube box；10-upper tube box；11-circulation pipe; 12- Condensate gauge bank of boiler steam.

a-the drain；b- port for vacuum system of the utilities system of the pharmaceutical factory；c, e- inlet of cooling water from the utilities system of the pharmaceutical factory；d, f- outlet of cooling water from the utilities system of the pharmaceutical factory; g- inlet of boiler steam from the pharmaceutical factory；h- outlet of boiler steam from the pharmaceutical factory；i- inlet of extraction liquids of traditional Chinese medicines；j-inlet of solid particles; k-vent.

can continuously run without scale formation and stable operation will be kept. At the same time, fluidized solid particles will scour the inner surface of the tube and thin or eliminate boundary layers close to inner walls of the tubes, and enhance heat transfer process.

The unit system was installed in a pharmaceutical factory in Tianjin, China. The heating steam, vacuum system, cooling water, power distribution and weak current system were all obtained from utilities system of the factory.

According to the generally required capacity of industry production of traditional Chinese medicine and the experience and knowledge on the lab-scale and pilot scale fluidized bed evaporators, the heat transfer area of the shell-and-tube heater in the fluidized bed evaporator unit with a vertical vapor-liquid-solid natural circulation boiling flow mode is designed to be 18.9 $m^2$, with the evaporation capacity of 1,100 - 1,400kg/h. There are two condensers of secondary steam. One is a plate type condenser with the heat transfer area of 49.5$m^2$, and the other is a shell-and-tube type condenser with the heat transfer area of 40.8$m^2$.

There are 81 tubes in the shell-and-tube heater, and they are made of stainless steel of 304 type. Its length is 1.95 m with outside diameter of 0.038 m and thickness of 0.002 m.

Operation parameters controlled in the fluidized bed evaporating concentration experiments of traditional Chinese medicines are shown in Table 1. For each operation parameter, the two numbers in Table 1 are the lowest and highest values or range taken in the experiments, and they don't represent the starting value and the end value of runs. They are also not random variations around mean value. The velocity of the circulation flow was not measured due to the limitations of the production conditions in the factory. However, it is generally higher than 0.5 m/s according to visual observation, using the CCD measuring technique, carried out in a transparent heating glass tube under similar operating conditions (Liu, et al., 2004).

In the experiments, alcohol and water extracts of Gengnian'an and other over ten kinds of traditional Chinese medicines were taken as the liquids to be concentrated. The concentration experiments were carried out in a semi-continuous mode or a fed-batch process. When the relative density of concentrated solution of thick liquor met the requirement of the product quality, the experiment stopped. The physical properties of the experimental liquids were estimated and representative results are shown in Table 2 (Liu, et al., 2005). The mass of water extraction liquids of Gengnian'an to be concentrated is about 24,000 kg. The mass of dilute solutions of other Chinese medicines is about 76,000 kg, including extraction liquids of alcohol solvent. Both alcohol and water extraction liquids were prepared by the multifunction extraction apparatus.

The cylindrical solid particles, both 0.003 m in length and diameter, with a density of 2200 kg/m$^3$ are made of physiologically inert material and the volume fraction of the solid particles filled in the heating tubes is 1-20%.

Table 1 Operation parameters in the fluidized bed evaporating concentration experiments.

| Operation parameters | Variations |
|---|---|
| Absolute pressure of steam, kPa | 115~400 |
| Absolute pressure in separator or vapor space, kPa | 10~90 |
| Temperature of liquor in separator, ℃ | 45~90 |
| Temperature drop of heat transfer or LMTD, ℃ | 20~78 |
| Fluctuation range of liquid level in separator, m | 0~0.3 |

Table 2 Physical properties of liquid phases of traditional Chinese medicines (Liu, et al., 2005).

| Extract | Temperature (℃) | Relative density (dimensionless) | Concentration of solid (w.t, %) | Viscosity, (m Pa·s) |
|---|---|---|---|---|
| Alcohol | 20 | 0.94 | 11.39 | 3.15 |
| | 79 | 1.35 | 89.01 | 18.53 |
| Water | 20 | 1.03 | 4.43 | 1.69 |
| | 88 | 1.35 | 94.07 | 20.68 |

Near the same methods and instruments were used to measure or calculate the experimental parameters in the industrial scale fluidized bed evaporation process (Liu, et al., 2005). These parameters include heating steam pressure, mass flow rate and temperature of the heating steam condensate, pressure and temperature of concentration liquor in separator, mass flow rate and temperature of the vapor in condenser, relative density and viscosity of the concentration liquor, volume fraction of the mixture of alcohol condensate, rate of heat transfer, overall heat-transfer coefficient and evaporation intensity of fluidized bed evaporator, etc. Among them, overall heat transfer coefficient ($K$, W/m$^2$K) and evaporation intensity ($U$, kg/h m$^2$) were calculated according to equations (1) and (2), respectively.

$$K = \frac{Q}{A\Delta t_m} = \frac{Q}{A(T_s - t_b)} \tag{1}$$

$$U = \frac{W}{A} \tag{2}$$

Where $t_b$ is boiling point of the extract, $K$; $T_s$ is the steam temperature at saturation pressure of $P_s$, $K$; $A$ is the heat transfer area, m$^2$; $W$ is the production capacity of an evaporator, which is defined as the number of kilograms of water vaporized per hour, *kg/h*; $Q$ is rate of heat transfer, W, which can be calculated by two methods: one is based on the mass rate of steam condensate and the other is on the basis of the mass rate of secondary steam or vapor condensate. Here, the second method was utilized for safety of design (Liu, et al., 2005). Generally, overall heat transfer coefficient $K$ is proportional to evaporation intensity $U$, and they are not independent of one another. However, different sides of operation characteristics of an evaporator can be obtained by analyzing the two parameters.

## RESULTS AND DISCUSSION

### Experimental results

**Zero fouling operation**

The experimental results of evaporating concentration of over 100,000 kg extraction liquids of Gengnian'an and more than ten other kinds of traditional Chinese medicines in the industrial scale fluidized bed evaporator show that in this industrial scale three-phase flow evaporator, the phenomenon of fouling or handling on the inner walls of heated tubes was eliminated and a stable and continuous three-phase flow concentration operation was also successfully achieved.

### Overall coefficient and evaporation intensity

**1. Compared to the typical industrial scale evaporator of vapor-liquid flow boiling**

Fig.2 show the relationship between the average of overall heat-transfer coefficient $K$ of the industrial scale fluidized bed evaporator and gauge pressure of the boiler steam when the extracts of traditional Chinese medicines were concentrated. Correspondingly, Fig.3 shows the evaporation intensity $U$ of the industrial scale fluidized bed evaporator and gauge pressure of the boiler steam when the liquids of traditional Chinese medicines were evaporated.

Compared to the ordinary industrial scale vapor-liquid flow boiling evaporation installation which is being used in the same factory under the approximate equipment structures and operating conditions, the three-phase fluidized bed evaporation equipment undergoes a higher overall heat transfer coefficient (Fig.2) and evaporation

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intensity (Fig.3). For the industrial scale vapor-liquid two-phase flow evaporator, overall coefficient and evaporation intensity are about 418.0 $W/m^2K$ and 23.2 kg/h $m^2$ for alcohol extract, respectively. Their symbols are '☆', as shown in Figs.2 and 3. Obviously, the points of these parameters are all on the bottom of the figures, which means that the values are the lowest compared to those of other systems. What is more, as expected, fouling on heating tube walls that often occurs in the ordinary vapor-liquid two-phase flow evaporation concentrator was not observed in this industrial scale three-phase flow evaporator.

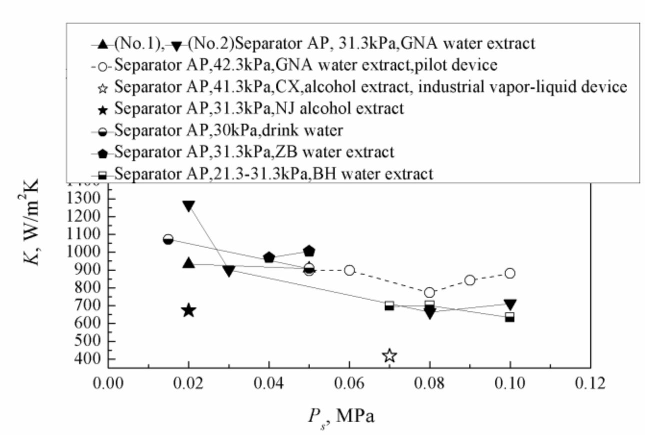


Fig.2 Overall heat-transfer coefficient $K$ of the industrial scale fluidized bed evaporator and gauge pressure of the boiler steam for different concentration liquids of traditional Chinese medicines.

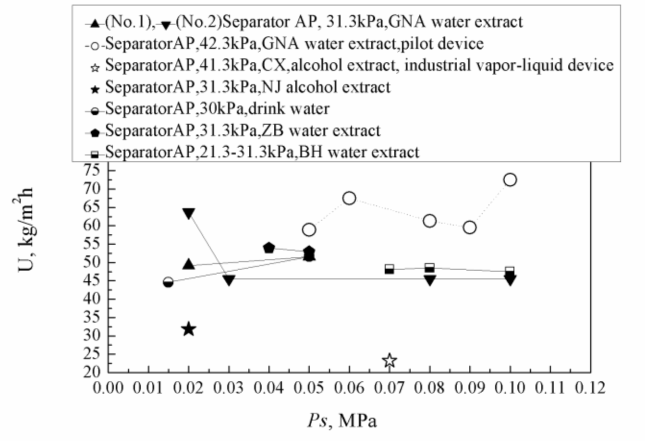


Fig.3 Evaporation intensity $U$ of the industrial scale fluidized bed evaporator and gauge pressure of the boiler steam for different concentration liquids of traditional Chinese medicines.

**2. Compared to the pilot scale fluidized bed evaporator**

We had developed a pilot scale fluidized bed evaporator, in which there are five heating tubes made of 316 stainless steel with outside diameter of 0.038 m, thickness of 0.0025 m and length of 2.9 m. Its heat transfer area is about 1.7$m^2$ and evaporation capacity 115 kg/h drink water (Liu et al., 2005). Compared to the test results of the pilot plant, the evaporation capacity of the industrial scale fluidized bed evaporation installation with heating area of 18.9 $m^2$ is in the range of 1100-1400kg/h drink water at approximate operating conditions with the absolute pressure of the separator of 21.3-31.3 kPa.

The overall heat transfer coefficient and the evaporation intensity of the pilot scale fluidized bed evaporator when the water extracts were concentrated are about 772.9-897.2 $W/m^2K$ and 58.9-72.5 $kg/m^2h$ (the symbol '○' in Figs.2 and 3) at the gauge pressure of the boiler steam from 0.05 to 0.1 MPa, respectively. While, the corresponding values of overall heat transfer coefficient and evaporation intensity for the industrial scale fluidized bed evaporator are about 662.9-711.7 $W/m^2K$ (Fig.2) and 45.5-48.5 $kg/m^2h$ (Fig.3), respectively, which are about 18-28% lower than those for the pilot scale fluidized bed evaporator. That is to say the experimental data of the industrial scale fluidized bed evaporator are generally about 72-82% of the pilot fluidized bed evaporator. This phenomenon is more obvious for the evaporation intensity values, as shown in Fig.3.

The experimental results of the overall heat transfer coefficient and evaporation intensity of the pilot test were not completely repeated in the industrial scale equipment and a factor of less than 1.0 on the overall heat transfer coefficient (about 82%) and evaporation intensity (about 72%) could be considered in the design of the industrial installations, which provides a useful guideline for the future industrial popularization and application of the fluidized bed evaporation process and installation.

In addition, the overall heat transfer coefficient and evaporation intensity of the alcohol extracts in the industrial scale fluidized bed evaporator are also all less than those of drink water extracts, which can be seen in Figs. 2 and 3 with symbol '★'.

**Discussion**

The difference of the overall coefficient or evaporation intensity between the pilot scale and industrial scale fluidized bed evaporators is considered to be scale-up effect. There are several reasons that result in the existence of the scale-up effect when enlarging the size of the fluidized bed evaporator compared to the data obtained from the pilot plant.

One main reason is that the distribution of fluidized solid particles in different heating tubes in the industrial scale installation is not as uniform as that of a single or only several heating tubes in a pilot plant. If the number of the heating tubes is too high, it is difficult to reach a uniform distribution of solid particles by vapor-liquid-solid boiling flow without a proper distributor of the particles.

Of course, the pressure of the heating steam, the vacuum degree of the separator, the characteristic of the traditional Chinese medicine extraction liquid, the solid particle characteristic and the height and diameter of the heating tube of the equipment also have certain impact on the performance of the fluidized bed evaporator.

Therefore, in the industrial design of this kind of evaporation installation, it is better to choose the overall heat transfer coefficient or the evaporation intensity of the fluidized bed evaporator with about 72% or 82 % discount of the pilot test data.

At present the industrial scale fluidized bed evaporator system has been successfully applied in the production of several traditional Chinese medicines after its verification of good manufacturing practice (GMP).

In addition, as mentioned above, the overall heat-transfer coefficient *K* or the evaporation intensity of alcohol extraction liquids is several hundreds lower than that of water extraction liquids, which is easy to understand because the heat-transfer coefficient of the organic liquids is often less than that of water solution.

It should be pointed out that the present equipment and operation parameters may not be their optimal values and parameter optimization is the further research work.

**Other examples of industrial applications**

Meanwhile, the fluidized bed evaporation technology and equipment developed here is also being applied to the other process industries. Some examples of industrial scale applications are only mentioned here due to the space limitations of the paper.

Fluidized bed evaporation technology was successfully used to solve the fouling problems met in the evaporation of inorganic salt solutions, such as magnesium chloride, calcium chloride, caustic soda, lithium hydroxide, lithium sulfate, phosphoric acid, mirabilite solutions, etc. In these applications, the natural circulation flow mode of the evaporator was often changed from the original forced circulation mode, and the circulation pump was omitted. After running a long time without stopping cleaning, no fouling layer was found on the tube walls, and the pressure drop of the fluidized bed evaporator didn’t increase too much (Li, 2017).

More fundamental studies and industrial applications on fluidized bed evaporation technology will be considered in the near future.

## CONCLUDINGS

The conclusions of this study are as follows.

1. The concentration experiments of extraction liquids of several traditional Chinese medicines in an industrial scale vapor-liquid-solid fluidized bed evaporator were carried out. Heat transfer enhancement and zero-fouling results similar to those of the pilot scale experiments were successfully obtained. Recent industrial scale applications in other processes were also obtained.
2. Fluidized bed evaporation technology is a physical method to online enhance heat transfer and to avoid fouling in the process industry. It can improve the heat transfer efficiency of heat exchanger, reducing coal consumption and environmental pollution, prolong the cleaning period, increase the actual production time, reduce labor intensity, reduce noise and improve operation environment, save energy, increase production, bring greater economic benefits, environmental benefits and social benefits.

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