Ultrasonic sprayer of liquid samples for atomic-emission microwave plasma analyzer

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Abstract. The design of an ultrasonic sprayer of aviation oils is proposed and its performance is investigated. Theoretical and experimental studies have been conducted to study the response of the spray tube when exposed to ultrasonic vibrations. Its resonance lengths are determined for given frequencies of ultrasonic vibrations. The aerosol flow parameters are estimated (length, flow diameter, droplet size), and the coefficient for the entry of particles of the analyzed metallic impurity in the sprayed liquid into the plasma is estimated.

1. Introduction

The atomic emission plasma complex has a significant difference from most spectral methods, which consists in a pulsed method of isolating and recording the analytical signal. The pulsed (scintillation) method of signal extraction allows obtaining information from one quantity of the sample on the particle size of the impurity being determined, their quantity, elemental composition of each particle and the mass fraction [1]. Express receipt of this information allowed us to successfully use this method to evaluate the technical condition of GTE oil systems of aircrafts and road transport by the parameters of wear particles [2-3].

The metrological characteristics of the plasma complex are largely determined by the system “spectrum excitation source – spray of liquid samples”.

When analyzing oil samples (measuring parameters of a metallic impurity), an original spectrum excitation source turned out to be the original design of a microwave plasma torch of a cyclone type [4], operating in air at atmospheric pressure.

To ensure the conditions of evaporation of metal particles, an oil sample must be introduced into the plasma in a spray state. At the same time, metal particles and oil droplets should be separated, i.e. metal particles should not be in a drop (shell) of oil. The resulting sol, consisting of oil droplets and individual wear metal particles, is passing to the plasma by a stream of carrier gas.

To reduce the effect of plasma temperature on the analytical signal, metal impurity particles must pass near the near-axial plasma zone. Ensuring that the particles move along the plasma near-axial zone can be achieved by a formed narrowly directed flow of sol, whose diameter is significantly less than the plasma diameter equal to about 6 mm.

For atomic emission and atomic absorption spectrometers, dozens of designs of pneumatic, ultrasonic, and other types of sprayers that transform a liquid into a cloud of sol with a particle size of
several microns have been developed. Analysis of the designs of various nozzles showed the absence of ready-made (industrial) developments suitable for use as a part of the microwave plasma complex.

The operating experience of the microwave plasma scintillation complex with different types and designs of nozzles revealed their shortcomings and allowed one to formulate a number of tasks (requirements), solution of which will allow optimizing the parameters of the fluid spraying and the conditions of its entry into plasma:

- maximum distribution of wear particles in size in serviceable aircraft engines accounts for 10-15 microns; therefore, in order to ensure the separation of oil droplets and metal particles, the sprayer must turn the oil into a sol with droplet sizes of the order of 10-15 μm;
- in scintillation measurements, the amount of wear particles entering the plasma and giving an analytical signal should approach 100%. This condition is provided by the spraying parameters, design of the burner and the flow rate of the carrier gas, which should not exceed 0.2 l/min [5];
- to ensure the conditions for the maximum entrance of the number of wear particles into the plasma, oil aerosol should be formed as a weakly diverging jet with a diameter of about 1.5-2 mm. Obviously, the larger the ratio between the diameters of the plasma and the sol jet, the greater the likelihood of the passage of wear metallic particles near the plasma axis, the higher the intensity of the spectral lines and the lower the error in determining the parameters in particles of the same size;
- operating rate of supplying sample oil to the sprayer is 100 μl/min. At this speed, the sprayer should ensure the formation of a uniform flow of the sol without breaks and pulsations, formation of large droplets larger than 20-30 μm in size is unacceptable;
- operating position of the sprayer is horizontal. The sprayer should be easily rinsed from the remnants of the previous sample, i.e. there should be no “pockets” and stagnant zones where wear particles could accumulate;
- in case of failure of the sprayer (interruption of the supply or spray tubes), operational possibility of repairing the sprayer within 10-15 min should be provided.

The purpose of the work was to develop the design of an ultrasonic sprayer for aviation oil samples that meets the above requirements.

2. The study of the response of the spray tube when exposed to transverse acoustic oscillations

The test of different types of oil sample sprayers in the microwave plasma complex showed that the ultrasonic sprayer described in [6] turned out to be the most acceptable one. In this design, liquid was sprayed due to ultrasonic transverse vibrations of a capillary tube with an internal diameter of about 0.9 mm. The sprayer made it possible to create an aerosol-generated stream with an oscillation frequency of ~ 22 kHz and a flow rate of the carrier gas of 0.2 l/min. At a frequency of more than 30 kHz, spraying stopped. The possibilities of creating a formed aerosol stream and a low flow rate of carrier gas were the basis for using it as a basis for further improvement.

The developed device for spraying viscous liquids (Fig. 1) is an ultrasonic oscillatory system made on 4 piezoceramic rings 2 mm thick, with an external and internal diameter of 10 mm and 5 mm, respectively. The nozzle, made of titanium BT3-1 (VT3-1), is attached to the acoustic waveguide with a threaded connection, with an oil supply and spray tubes, the internal diameter of which is 0.75 mm and the external diameter of 1.05 mm. Along the axis of the ultrasonic oscillatory system and the spray nozzle there is a channel for supplying the carrier gas to the spray nozzle.

To assess the character of the oscillations of the spray tube when applying transverse ultrasonic vibrations to it, a mathematical simulation of the process was performed, where the spray tube was represented as an oscillating hollow rod with one free end.
Figure 1. A scheme of the device for spraying samples of aviation oils.

The equation describing the transverse oscillations of a rod of length \( l \) with a free end has the following form [7]:

\[
\frac{\partial^2 y}{\partial t^2} + a^2 \frac{\partial^4 y}{\partial x^4} = 0, \quad a^2 = \frac{EJ}{\rho S} \tag{1}
\]

where \( E \) is the modulus of elasticity of the rod material, \( \rho \) is the material density, \( S \) is the cross-sectional area of the rod, \( J \) is the moment of inertia of the section of the rod about its horizontal axis. For tubes with internal \( d_1 \) and external \( d_2 \) diameters (needle):

\[
J = \frac{\pi}{64} (d_2^4 - d_1^4). \tag{2}
\]

Let us find a solution to equation (1) in the form of separated variables:

\[
y(t, x) = Y(x) T(t). \tag{3}
\]

Substituting it into the equation, we have:

\[
\frac{T''(t)}{a^2 T(t)} = -\lambda \frac{Y^{(4)}(x)}{Y(x)} = -\lambda. \tag{4}
\]

Function \( Y(x) \) satisfies the equation:

\[
Y^{(4)} - \lambda Y = 0. \tag{5}
\]

A general solution of this equation is:

\[
Y(x) = A \cosh \alpha x + B \sinh \alpha x + C \cos \alpha x + D \sin \alpha x, \quad \alpha = \sqrt[4]{\lambda}. \tag{6}
\]

Boundary conditions in case of forced fluctuations are the following. Free end conditions:

\[
Y'(l) = 0, \quad Y'''(l) = 0. \tag{7}
\]

The conditions at the clamped end are determined by the function \( f(t) \); we consider it harmonic:

\[
Y'(0) = 0; \tag{8}
\]
\[ y(t,0) = f(t) = F \cos \gamma t \]. 

Conditions (7) - (8) make the solution (6) with the only one arbitrary coefficient A:

\[ B = -A \frac{(ch \alpha l \sin \alpha l + sh \alpha l \cos \alpha l)}{(1 + ch \alpha l ch \alpha l + sh \alpha l \sin \alpha l)} \]  

(10)

\[ C = A \frac{(1 + ch \alpha l ch \alpha l - sh \alpha l \sin \alpha l)}{(1 + ch \alpha l ch \alpha l + sh \alpha l \sin \alpha l)} \]  

(11)

\[ D = -B \]. 

(12)

In condition (9) we need the value \( Y(0) \). In view of (10), it is equal to:

\[ Y(0) = 2A \frac{(1 + ch \alpha l ch \alpha l)}{(1 + ch \alpha l ch \alpha l + sh \alpha l \sin \alpha l)} \]  

(13)

The function \( T(t) \) satisfies the equation:

\[ T'' + \lambda^2 T = 0 \]. 

(14)

A general solution:

\[ T(t) = c \cos \omega t + d \sin \omega t, \quad \omega = a \sqrt{\lambda} \]. 

(15)

Finally, we impose the condition (9): the clamped end of the rod moves according to the harmonic law. It gives the following ratio:

\[ (c \cos \omega t + d \sin \omega t) Y(0) = F \cos \gamma t \]  

(16)

whence it follows that:

\[ \omega = \gamma, \quad \lambda = \gamma^2 / a^2 \]  

(17)

\[ c = F / Y(0), \quad d = 0 \]. 

(18)

Thus, the solution that corresponds to steady-state forced oscillations is completely determined.

In the equations mentioned above, natural frequencies lead to an infinite value \( Y(0) \) (14), and are determined by the equation:

\[ 1 + ch \alpha l ch \alpha l + sh \alpha l \sin \alpha l = 0 \]  

(19)

Figure 2 shows the results of calculations of the amplitude of oscillations of the spray tube end depending on its length, which shows that when the frequency of the driving force coincides with the eigenfrequencies of the rod, a resonance occurs, and the amplitude of the oscillations formally goes to infinity. In other words, at a fixed frequency of ultrasonic oscillations, it is possible to choose the length of the spray tube when there is a significant increase in the amplitude of oscillations of the free end in comparison with the fixed one.

Figure 3 shows the results of calculations of the resonant frequency of oscillation of the spray tube of various lengths, with an external diameter of 1.05 mm and an internal diameter of 0.75 mm (square points). The same figure shows the experimentally obtained data on the frequency ranges of the sprayer stable operation (stable spraying) for spray tubes with the length of 25.8 mm, 26.5 mm and 27 mm.
Figure 2. Calculated values of the amplitude of oscillations of the spray tube end, depending on its length, where $Y(0)$ – Conditions at the clamped end, dimensionless quantity, $L$ – the length of the spray tube.

Figure 3. Calculation results and experimental data on the resonance of the spray tubes of various lengths.

Figure 3 shows a satisfactory coincidence between the calculated values and the experiment. Thus, with a tube length of 27 mm, the resonant frequency was 22 kHz, while the experiment showed a range of 19.8–20.4 kHz, i.e. the discrepancy was about 10%.

The results shown in Figure 3 also show that spraying at the selected tube length occurs in a rather narrow frequency range not exceeding 1.0 kHz. This imposes relevant requirements on the stability of the ultrasonic generator.

It should be added that the data obtained for the frequency ranges at which spraying occurs refer to tubes whose ends have a perpendicular cut to its axis. In case when the free end of the tube has a cut at an angle of 30°, the range of stable spraying increases, and is at least 1.5 kHz.

Figure 4. A video capture frame of the sprayer operation. The tube with a perpendicular cut. External tube diameter is 1.05 mm.

Figure 4 shows the sprayer operation at a generator frequency of 21.01 kHz and a spray tube length of 26.5 mm with a perpendicular cut under the following experimental conditions.

Aviation oil MC-8П (MS-8P) (viscosity 8 cSt) with the help of a special device was evenly supplied at a speed of 100 μl/min to the sprayer through the oil supply tube. Through the feed gas supply nozzle (Figure 1), air was supplied at a rate of 0.2 l/min. For visual evaluation of the character of spraying (stability, size of the formed aerosol stream), video of the stream, illuminated by a laser beam defocused to a diameter of 7 mm, was used.
Figure 4 shows the absence of breaks (pulsations) of the jet, the flow is slightly divergent, located horizontally, the diameter of the jet at a distance of 30 mm from the tube end does not exceed 2 mm. Thus, the above tasks of optimizing the design of the sprayer (ease of mounting and removing of the spray nozzle from the waveguide, size of the aerosol flow, horizontal arrangement of the spray tube, absence of pulsations and the uniformity of spraying) are solved.

3. Measurement of dispersed oil droplet size
The determination of the dispersed composition of a liquid during ultrasonic spraying has been described in many papers. There is expensive equipment for complex measurement of parameters: measurement of the sol flow rate, particle direction of movement and size, such as PhaseDopplerAnemometry (PDA). There are simple methods and equipment based on applying droplets of a sol on a substrate with the subsequent measurement of the dimensions using a microscope [8].

For various reasons, these methods could not be used to measure the size of droplets of sprayed aviation oil. In this regard, we have developed a method, the essence of which is as follows (Figure 5):

![Figure 5. A block diagram of the installation for measuring the size of the sprayed oil droplets.](image)

Oil is sprayed into the sprayer through the inlet pipe and sprayed into the air. At a distance of 20 mm from the end of the spray tube (the point of entry of the aerosol into the plasma is the lens of the “Micromed-1” microscope with a recording video system with a 100-fold magnification of the system). It is also possible to use 40-fold magnification. The center of the flow of aerosol is in the focal plane of the microscope objective.

On the opposite side of the spray tube, an oil sol stream is visualized with the help of a laser, the beam of which is turned by a cylindrical lens into a plane. Laser-illuminated droplets are recorded by the microscope video system in the form of individual tracks, where the track width is the drop size, and its length is proportional to the exposure time and the speed of the drop. The width of the track, taking into account the image of the dimensional scale at the same magnification, is converted to the particle diameter.

With a 100-fold increase in the system, the depth of field is small, so only tracks with a sharp boundaries outline are selected for calculation in the photo (video).

To verify the accuracy of particle size measurements across track widths, the method was tested on particles of known sizes in two different ways.

A powder membrane sprayer was connected to the sprayer supply tube, according to the type described in [9]. A calibrated quartz powder with a known particle size distribution was loaded into the sprayer. Carrier gas was fed into the spray chamber at a rate of 0.2 l/min, a vibrator was turned on, and the mixture was blown through the exhaust pipe with a diameter of 1.05 mm. The particles were
illuminated by a laser beam and recorded by a microscope video system. According to the obtained tracks, particle size was calculated, size distribution was plotted (Figure 6a).

To further confirm the obtained data on the distribution of quartz particles, their size was measured in another way, which was as follows. With the help of a camera, pictures of particles on a microscope substrate were taken and their image was recalculated into a size using a special program (Figure 6b).

![Figure 6. Size distribution of SiO2 particles measured: a) in the gas stream along the tracks; b) in size of particles on the photo with high magnification under a microscope.](image)

It should be noted that the distribution of particles plotted according to measurements of the track width is slightly shifted to the right, the maximum of the distribution falls on 25-30 microns. This can be done in such a way that the visible size of some tracks of particles extending below or above the focal plane of the lens increases, giving, respectively, an increased particle size.

To measure the size of the sprayed oil droplets, an ultrasonic oil spray was installed instead of the powder one, the aerosol flow was directed to the focal plane of the microscope objective, a gauge was measured, and then, the sprayed oil droplets were measured.

![Figure 7. Distribution of oil droplets by size: a) tracks of oil droplets, b) histogram of the distribution of oil droplets by size.](image)

Figure 7 shows the image of the tracks of MC-8P (MS-8P) oil droplet in a stream illuminated by a laser, and their size distribution.

Thus, the results, shown in Figure 7, show that when applying ultrasonic oscillations with a frequency of 22 kHz to the spray tube with an internal diameter of 0.75 mm, the maximum size of the droplets practically does not exceed 20-30 μm. The maximum distribution is 10 μm.

For a reliable evaluation of technical condition of the friction units washed by the oil according to the results of scintillation measurements, the entire range of particle sizes in the oil sample should be
considered. In other words, in the ideal case, 100% of the particles in the analyzed sample should enter the plasma and produce analytical signal.

Since when a sample is introduced into a cyclone-type microwave plasma torch, some of the oil droplets do not enter the plasma, but settle on the surface of the plasma-forming chamber, flowing through the drainage hole into the drain, it becomes necessary to estimate the coefficient of sample entry. For the experimental evaluation of the coefficient of wear particles entering the excitation source of the cyclone-type spectra, an oil sample with a known composition and a constant composition was prepared. As such an 8-element oil standard sample of the SOCHPI GTE UNIIM (GSO 10696-2015) with a copper content of 1.5 g/t was used. The volume of sprayed oil, the volume of the resulting drain, the number of particles registered in the analysis of the initial samples and the analysis of the total volume of the drain were measured.

After analysis of 28 samples of SOCHPI (28 ml of oil in total), volume of drain was 7.5 ml. The results of measurements are shown in Tables 1 and 2.

![Table 1. The results of measuring the number of particles in the initial SOCHPI.](image)

| No./elem | Al  | Cr  | Ni  | Mg  | Fe  | Cu  | Ag  | V  |
|----------|-----|-----|-----|-----|-----|-----|-----|----|
| The number of registered particles in 1ml of the CO Sochpi sample | 124 | 330 | 39 | 1214 | 940 | 2283 | 158 | 1 |

![Table 2. The results of measuring the number of particles in the sample drain.](image)

| No./elem | Al  | Cr  | Ni  | Mg  | Fe  | Cu  | Ag  | V  |
|----------|-----|-----|-----|-----|-----|-----|-----|----|
| The number of registered particles in 1ml plum | 18 | 11 | 4 | 54 | 42 | 82 | 5 | 0 |

The coefficient of entrance of metal particles is estimated as the difference of 100% and the ratio of the average number of particles registered in the “drain sample” to the average number of particles registered in the SOCNI sample, normalized to 1:

\[
k = 1 - \left( \frac{\overline{N}_{dr.}}{\overline{N}_{in.} + \overline{N}_{dr.}} \right)
\]

where, \( k \) – the coefficient of entry of particles in the spectrum excitation source (SES), \( \overline{N}_{dr.} \) – average number of particles registered in the “drain sample”, \( \overline{N}_{in.} \) – average number of particles registered in the initial sample of the SOCHPI.

Results for different elements are summarized in Table 3.

![Table 3. The coefficient of the entry of metal particles into the source of excitation of spectra for different elements.](image)

| k | Al  | Cr  | Ni  | Mg  | Fe  | Cu  | Ag  | average |
|---|-----|-----|-----|-----|-----|-----|-----|---------|
| k1 | 87.53 | 96.69 | 90.98 | 95.74 | 95.72 | 96.53 | 96.67 | 94.27 |

Based on the data given in Tables 1-3, a number of conclusions can be made.

1. Drain volume was 26.8% of the volume of the sprayed samples. However, there was practically no wear metal particles in the drain. Due to inertia, denser metal particles overcome the temperature barrier and enter the plasma, while less dense ones (almost an order of magnitude) oil droplets are partially reflected from it and fall on the walls of the discharge chamber.

2. The share of wear particles that fell into the drain from the total number of them in the operating oil samples varied randomly from 1 to 4%. Considering the large (about 100% and higher) component
of the random error of counting rare events (particles in the drain), the systematic error associated with the loss of particles in the drain can be considered insignificant compared to the total random error of their count. Therefore, the number of registered pulses in a scintillation measurement from an analytical sample of 1 cm$^3$ is indeed the number of wear particles per unit volume of the analytical sample.

4. Conclusion
The results of the tests allow making the following conclusions:

  1. When applying transverse ultrasonic oscillations of a fixed frequency to the spray tube, the tube length at which resonance occurs can always be chosen. Herewith, the amplitude of the free end can significantly exceed the amplitude of the fixed end, i.e. oscillations increase.

  2. The results of theoretical and experimental evaluation have shown that with a fixed frequency of ultrasonic oscillations, spraying with the selected tube length, is observed in a very narrow frequency range, not exceeding 1 kHz.

  3. A method for measuring the size of liquid droplets, which are established with ultrasonic oscillation frequencies of about 22 kHz and spray tube sizes of 1.05 mm and 0.75 mm (external and internal diameter, respectively), has been developed. The proposed sprayer design provides an average droplet size of 10 μm when spraying MC-8II (MS-8P) aviation lubricant oil.

  4. The sprayer of this design allows forming an extended stream of aerosol without breaks and pulsations. The diameter of the flow at a distance of 30 mm from the end of the spray tube does not exceed 2 mm.

  5. The sprayer has no “pockets” and stagnant zones and is easily washed from the remnants of the previous sample. The threaded connection of the sprayer with the acoustic waveguide makes it easy (in a few minutes) to change the spray nozzle in case of its replacement or breakage.

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