REMOTE DETECTION OF AMMONIA LEAKAGE UNDER INDOOR CONDITIONS

R.R. Agishev, L.R. Aibatov, V.A. Vlasov, and R.K. Sagdiev

A.N. Tupolev State Technical University, Kazan'

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In this paper we discuss remote devices to monitor automatically ammonia leakage under indoor conditions. The devices are economical and easy to use; their operation is based on methods of spectroscopic analysis.

INTRODUCTION

In some practices involving operations with dangerous gases, the risk of emergency is run. To detect a leakage of such gases timely, continuous routine monitoring is needed. Currently available approaches are based either on laboratory analysis of air samples or on various methods of monitoring *in situ*. The former ones are characterized by a long time required for sampling, transporting samples, and conducting measurements. Therefore, they cannot provide for a prompt detection. The latter ones ensure continuous automatic monitoring only at a few preset sites. That is why it is urgent to develop remote devices capable to detect gas leakage under both indoor and outdoor conditions in a real time mode.

Ammonia vapor falls in the group of major pollutants to be monitored continuously.

1. IR SPECTRAL REGIONS SUITABLE FOR AMMONIA DETECTION

The list of most intense absorption lines of ammonia in the region from 1.5 to 12.6 μm is given in the Table I along with the corresponding absorption cross sections.

TABLE I.

Wavelength, μm	Absorption cross section, cm^2	Reference
12.66	$2.35 \cdot 10^{-19}$	1
10.58	$2.15 \cdot 10^{-17}$	1, 2
6.13	$4.1 \cdot 10^{-18}$	1
5.75	$4.17 \cdot 10^{-20}$	1
2.0 ... 3.0	$1.4 \cdot 10^{-19}$	2
near 1.5	$(1 ... 2) \cdot 10^{-21}$	3

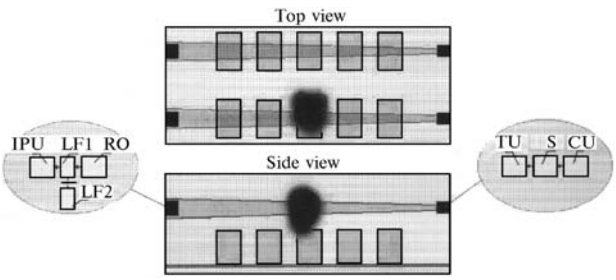
The spectral region from 1.5 to 3.0 μm is suitable for development of inexpensive and convenient devices with the radiation source and receiver not requiring cryogenic cooling.

Spectral lines near 1.5 μm are very promising for analytical applications, although being weaker than others. In this case, A^3b^5 diode lasers (DLs) operating in the near IR at room temperatures can be used.

As to the region from 2.0 to 3.0 μm, it is worthwhile to use broad-band sources ("traditional" thermal sources, light-emitting diodes) and light filters for cutting-off ammonia absorption bands in a receiving channel.

The method of differential absorption is convenient for the development of economical devices, because it combines acceptable sensitivity with the ease of technical implementation.[1,2]

A possible design and structure scheme of a remote ammonia detector are shown in Fig. 1.



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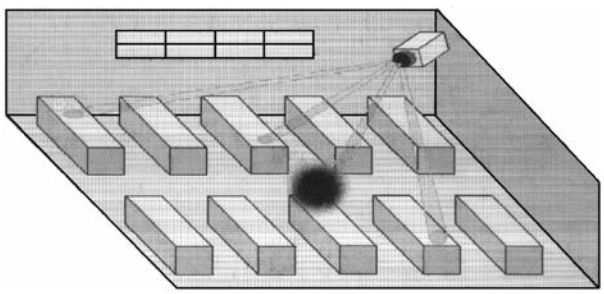

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FIG. 1. Design and block diagram of the opto-electronic system for monitoring ammonia under indoor conditions.

2. ANALYSIS OF POWER CHARACTERISTICS OF A SYSTEM BASED ON LASER DIODES

In the differential absorption method, it is commonly accepted to estimate power of a signal coming to the photodetector as follows:

$$P = G\, P_{out} \exp(-\alpha L - \alpha_{atm} R)\, t, \qquad (1)$$

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where R is the path-length; L is the linear size of a gas cloud along the sounding path; α is the ammonia absorption coefficient; t is the optics transmission; G is the geometry factor, $G = (D_r/D_{sp})^2$, D_r is the diameter of the receiving optics, D_{sp} is the diameter of the radiation spot at the receiver; α_{atm} is the atmospheric extinction coefficient. For power estimates under conditions of a thin haze, we took $\alpha_{atm} = 10^{-1}$ km^{-1} in the wavelength range about $\lambda = 1.5$ μm.

The power of signal which bears information about ammonia is estimated by a difference in the power corresponding to an edge and center of an absorption line:

$$P_v = G\, P_{out} \exp(-\alpha_2 L - \alpha_{atm} R)\, t\, [1 - \exp(-\Delta\alpha L)]. \quad (2)$$

Here $\Delta\alpha = \alpha_1 - \alpha_2$, where α_1 and α_2 are the ammonia absorption coefficients at the center and edge of an absorption line, respectively.

According to Ref. 4, the reduced value of the ammonia absorption coefficient at the center of the line near 1.5 μm is the following:

$$\alpha_1/N \approx 2.5\cdot10^{-8}\ \text{cm}^{-1}\cdot\text{mg}^{-1}\cdot\text{m}^3,$$

where N is the ammonia density, in mg/m^3, and $\Delta\alpha/N = 1.1\cdot10^{-8}$ cm^{-1}·mg^{-1}·m^3.

The threshold power for the method of direct photodetection is equal to

$$P_t = (S\,\Delta F)^{1/2}/D^*, \quad (3)$$

where D^* is the photodetector's detectability; S is the area of its photosensitive surface; ΔF is the passband of the receiving channel. Modern PbS photoresistors have $D^* = (0.3 \ldots 1.0)\times10^{11}$ cm·Hz$^{1/2}$/W in the spectral range $\lambda = 1.5 \ldots 3.0$ μm at the room temperature. They may be designed to have the photosensitive area from several fractions of square millimeter to several square millimeters. Thus, for $S = 0.25\times0.25$ mm^2, $D^* = 10^{11}$ cm·Hz$^{1/2}$/W, and $\Delta F = 100$ Hz, we have $P_t = 2.5\cdot10^{-12}$ W.

When operating against a bright background, one should take into account the background of the daytime sky P_b. According to Ref. 5

$$P_b = \pi\, B_\lambda\, \Delta\lambda\, S_r\, t\, \theta^2/4, \quad (4)$$

where B_λ is the spectral brightness of the sky background; $\Delta\lambda$ is the passband of the interference filter; θ is the plane angle of the receiver field of view; S_r is the receiving lens area. On the assumption that $B_\lambda = 0.1$ W/(m^2·sr·μm), $\Delta\lambda = 0.012$ μm, and $\theta = 3$ mrad, we obtain $P_b \approx 2\cdot10^{-10}$ W, that is, under real conditions the photodetector sensitivity is restricted by the background. In this case, the minimal detectable ammonia concentration N_{min} can be found from the condition $P_{c\,min} = P_b$.

Using an InGaAsP/InP diode laser with the mean output power of 5 mW and the beam divergence about 3 ... 5 mrad at the path-length $R = 30$ m and linear extent of the ammonia cloud $L = 1$ m, from Eqs. (1)–(4) we can find that $N_{min} = 0.28$ mg/m^3, what 100 times exceeds the human sense of smell.

The functional diagram of the DL-based device is shown in Fig. 2.

Below we explain briefly the device operation according to this diagram. The radiation wavelength of a semiconductor laser (SCL) is modulated harmonically in the region of a gas absorption band with the frequency f_{mod}. The transmission channel tunes the output radiation wavelength, stabilizes the mean value of the wavelength, and suppresses parasitic oscillations of the SCL power. A fraction of the radiation passed through the volume under study, comes to the receiving optics, which transforms an optical signal into an electric one and amplifies it. Then the signal at the frequency f_{mod}, which bears information about the pollutant concentration, is separated out and normalized to the mean power of incident radiation. A micro-computer is used to calculate the concentration value.

3. CHARACTERISTICS OF THE AMMONIA CONCENTRATION METER BASED ON IR LIGHT-EMITTING DIODES

Modern IR light-emitting diodes possess pulse power p_p of several tens of mW at a pulse duration on the order of 100 ns. Light-emitting diodes emit radiation within a band, whose width $\Delta\lambda_{rad}$ is a few fractions of micrometer. To separate out narrower spectral bands, narrow-band interference filters should be utilized in the receiving channel. Rather typical and relatively cheap filters have the minimal passband $\Delta\lambda_f \approx (0.005 \ldots 0.01)\times\lambda_0$, where λ_0 is the wavelength corresponding to the filter transmission maximum. In this case, the expressions for power of received signals should be multiplied by the factor

$$\eta_\lambda = \Delta\lambda_f/\Delta\lambda_{rad},$$

where η_λ is the spectral efficiency of the radiation source.

The ammonia absorption spectrum in the region from 1.9 to 3 μm has the lines with the maximum absorption cross section $\sigma_{max} \approx 1.4\cdot10^{-19}$ cm^2. This allows one to select neighboring spectral ranges with the width $\Delta\lambda_f$ in such a way that the mean absorption cross section in one spectral range is close to the maximum value, while in the another range it may be half as small. Then the following estimate can be obtained for neighboring bands: $\Delta\alpha/N \approx 2.4\cdot10^{-6}$ cm^{-1}·mg^{-1}·m^3.

If a light-emitting diode operates in a pulsed mode, the receiving channel should have the passband $\Delta F = 10^7$ Hz. Then the threshold power of an optical signal at the photodetector output, according to Eq. (3), increases up to $7.9\cdot10^{-10}$ W, what exceeds the background level given by Eq. (4). In this case, the device sensitivity can be found from the condition $P_{c\,min} = P_t$. For the LED21 light-emitting diode ($\lambda \sim 2$ μm) having the pulse power of 60 mW (the divergence of a beam from the light-emitting diode transmitter $\theta = 1°$), the threshold sensitivity of the device is $N_t = 0.05$ mg/m^3.

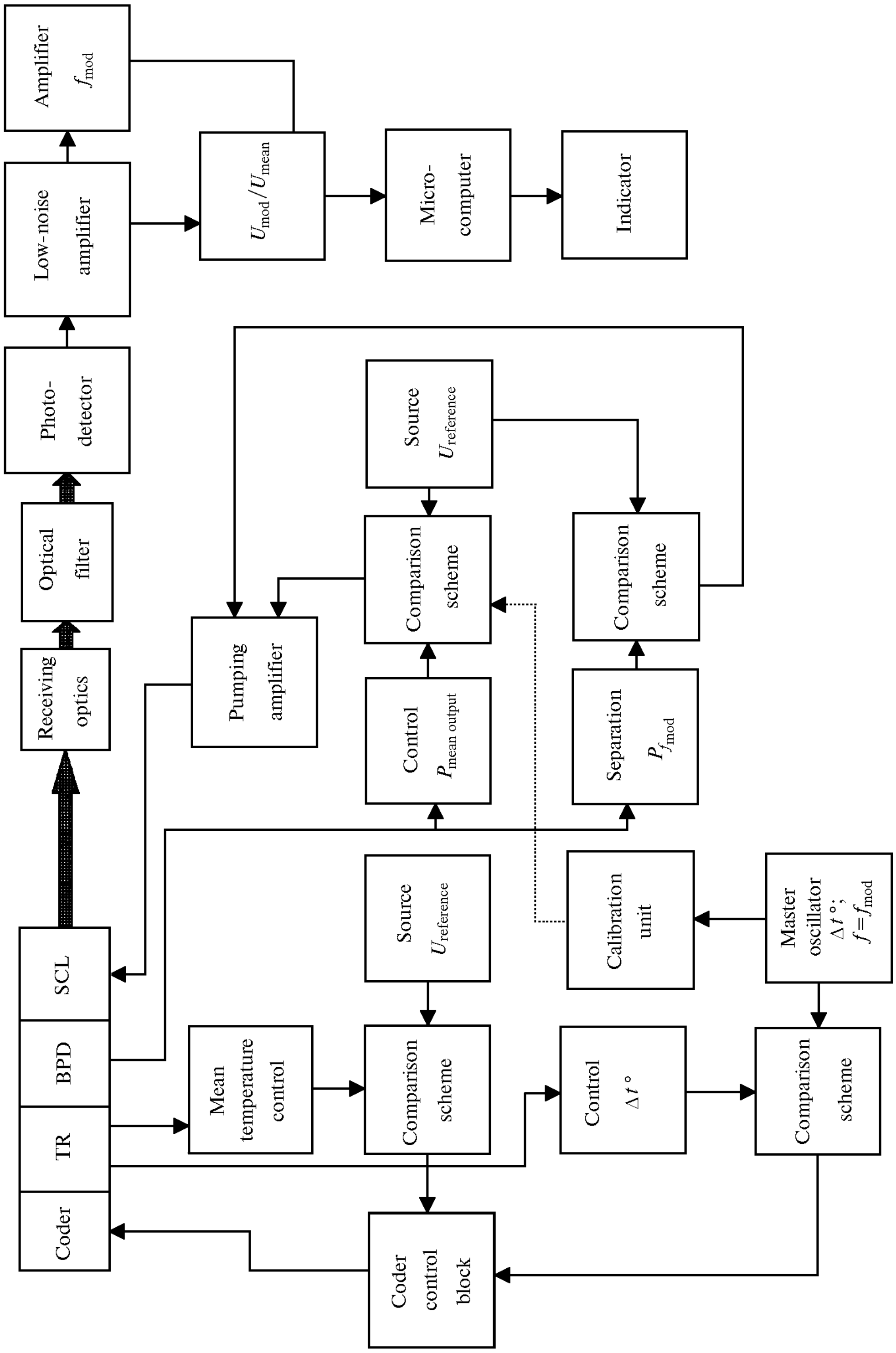

FIG. 2. Functional diagram of the ammonia leakage detector: thermoelectric cooler (cooler), thermoresistor (TR), built-in photodetector (BPD), semiconductor laser (SCL).

When the light-emitting diode operates in the cw mode, the output power is about 1.5 mW. In this case, the receiving channel may have the passband width $\Delta F = 100$ Hz, and the photoreceiver sensitivity is restricted by the background noise (4). The device sensitivity in this case drops down to $N_t = 0.07$ mg/m^3, what is also quite sufficient for practical use.

The functional scheme of the device based on the light-emitting diodes, which implements the usual two-channel scheme, is shown in Fig. 3. The broad band radiation from a light-emitting diode, upon passing through the studied volume, comes to the receiving optics. The filter LF1 is tuned to the center of the ammonia absorption band, while the filter LF2 is tuned to an edge of this band. The differential amplifier subtracts two signals. The pollutant concentration is calculated in the indicator and processing unit.

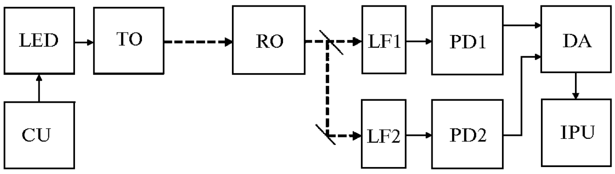

FIG. 3. Functional diagram of the gas analyzer based on the light-emitting diodes: control unit (CU), light–emitting diode (LED), transmitting optics (TO), receiving optics (RO), filter (LF), photodetector (PD), differential amplifier (DA), indicator and processing unit (IPU).

4. ESTIMATE OF POWER CHARACTERISTICS OF A THERMAL SOURCE OF RADIATION

In the spectral region from 2 to 4 μm, thermal sources of radiation can also be used. When a lamp heating element warms up to 1000 K, radiation intensity is maximum at 3 μm. Approximating the spectral characteristic of the thermal source by the uniform distribution law with the width $\Delta\lambda_{rad} = 3$ μm and assuming the filter passband $\Delta\lambda_f = 0.01$ μm, we can find the spectral efficiency of the source $\eta_\lambda = \Delta\lambda_f/\Delta\lambda_{rad} = 3\cdot10^{-3}$. At the source power of 300 W, the effective power $p_{eff} = 900$ mW falls within the filter passband. This is hundreds times higher than the power of the light-emitting diodes considered above. Therefore, the thermal source can provide better concentration sensitivity of the device, and it is suitable for analysis of different pollutants due to a wide spectral range of the emitted radiation. In this case, to change a pollutant to be analyzed, one needs simply to change a filter.

5. ESTIMATE OF CHARACTERISTICS OF AN IR LFM LIDAR FOR REMOTE DETECTION OF AMMONIA LEAKAGE

Continuous lidars with linear frequency modulation of optical beam intensity provide quite sufficient spatial resolution along paths at a low mean power (several mW) of the emitting laser.[6] In this case, the method of differential absorption is used, and the lidar equation takes the following form:

$$P = G\ E\ c\ S_r\ \beta\ \exp(-2\alpha\ R)\ t/R^2,$$

where c is the speed of light; e is the laser output power; $e = p_{rad}\ t_{ac}$, p_{rad} is the laser radiation power in a cw mode, t_{ac} is the time of received signal accumulation; β is the volume backscattering coefficient. Approximating the scattering phase functions at an angle π as[7]

$$i_\pi = 0.01\ \alpha^{-0.43},$$

we can estimate the equivalent signal power at the photodetector input as:

$$P = 0.01G\ E\ c\ G_r\ \alpha^{0.57}\ \exp(-2\alpha\ R)\ t/R^2.$$

In the spectral range near λ = 1.5 μm, the above-mentioned photoresistors can be used as a photosensitive element. The PMTs allowing the operation in the heterodyne mode do not work here. The receiver includes a photoresistor, which transforms the optical LFM backscattered signal into the electric signal. Then the signal comes to a mixer, and the LFM heterodyne signal comes to the mixer as well. The mixer produces a difference-frequency signal, which is proportional to the distance to studied sections of the path.[6,7,9]

To provide for the required spatial resolution (ΔR of several meters), the modulation frequency band $\Delta F \approx 10 \ldots 25$ MHz is required, as well as the same transmission band of the photodetector. Then the threshold sensitivity is $P_t = 8\cdot10^{-10}$ W for the above-mentioned photoresistors at $\Delta F = 10$ MHz.

The above-considered InGaAsP/InP diode laser can be used as a radiation source for the lidar. However, with regard for the 100% modulation, the mean power should be taken equal to 2.5 mW. In this case, for a 200-m long path and the signal accumulation time about $T_{ac} = 1$ ms, at the ammonia concentration $N = 1$ mg/m^3 the ratio of the received signal power to the threshold power can be estimated as $P_{sig}/P_t \approx 2$.

CONCLUSION

Thus, the analysis shows that application of the relatively simple technical solutions (with regard for the assumptions made) allows development of inexpensive devices for remote monitoring of ammonia and other gases. Such devices could allow detection of ammonia leakage, possess good practical characteristics, and provide for the required spatial resolution.

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