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The effect of elevated water sample temperature on the performance of a custom-developed colorimetric arsenic sensor

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Abstract

A custom-developed colorimetric device is presented for arsenic ion detection in drinking water. The device utilizes standard reagents, which are used in arsenic field-testing kits, in its custom microfluidic system. The response for arsenic concentrations is linear in the 5-20 µg/l (ppb) range, thus our device is capable to distinguish drinkable water based on the World Health Organization (WHO) guidelines1 limit of 10 µg/l. The effect of elevated water sample temperature on the kinetics and performance of the device is investigated in order to reduce the required time for the measurements. Stable and reliable arsenic concentration values were obtained after 25 min at 50 ºC water sample temperature.

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Keywords: Arsenic; Colorimetry; Microfluidics

1. Introduction

The arsenic contamination of drinking and irrigation water systems is one of the major environmental challenges of our century. Besides its direct toxicity, the mutagenic and carcinogenic potential of arsenic upon prolonged exposure is also reported. While there are several reliable methods for arsenic monitoring [1], the available techniques require expensive and complex instrumentation with highly skilled staff. There would clearly be need for simpler, portable (preferably handheld) devices for on field testing and monitoring [2]. Colorimetric methods using various reagents [3] are offering a simple an economic way to determine arsenic concentrations and some of these are already available as field test kits (Fig. 1). However, the operation of such kits could still be complicated for ordinary untrained field users,

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so the development an easy to use device with built-in, automatic evaluation could be a valuable asset, especially for developing countries, where the arsenic contamination of water systems is the most severe around the world.

2. Materials and Methods

In cooperation with S-Metalltech 98 Co. (Hungary) our department developed a small device (Fig. 2), which utilizes colorimetric detection and the standard reagents used by an on filed arsenic testing kit, namely the Hach® EZ Arsenic Test Kit [4] (Fig. 1/A), integrated into a microfluidic setup (Fig. 3).

In the method used by Hach®, hydrogen sulfide is first oxidized to sulfate to prevent interference, then the oxidizing environment is neutralized. Next, sulfamic acid and powdered zinc react to create strong reducing conditions in which inorganic arsenic is reduced to arsine gas. The arsine gas then reacts with mercuric bromide, impregnated onto a test paper to form mixed arsenic/mercury halogenides (e.g. AsH_2HgBr). The mixed halogenides discolor the test strip to a degree proportional to the concentration of arsenic in the sample. The color change is from white to yellow to tan to brown.

Our developed device calculates the concentration of arsenic in the sample by determining an RGB color value for the digitized picture of the test strip and then the relative proportion of the blue component is computed. Red, Blue and Green components are calculated by summarizing pixel color values on the localized discolored area. In order to define the relation between the relative blue component of the calculated RGB value and concentration of arsenic in the sample, an experimental calibration measurement was carried out. The calibration measurement was performed in triplicates by using the original reaction vessel and original reagents of the Hach® EZ Arsenic Test Kit. The field kit with its reagents, the reference color series and a photograph of the results is shown in Fig. 1.
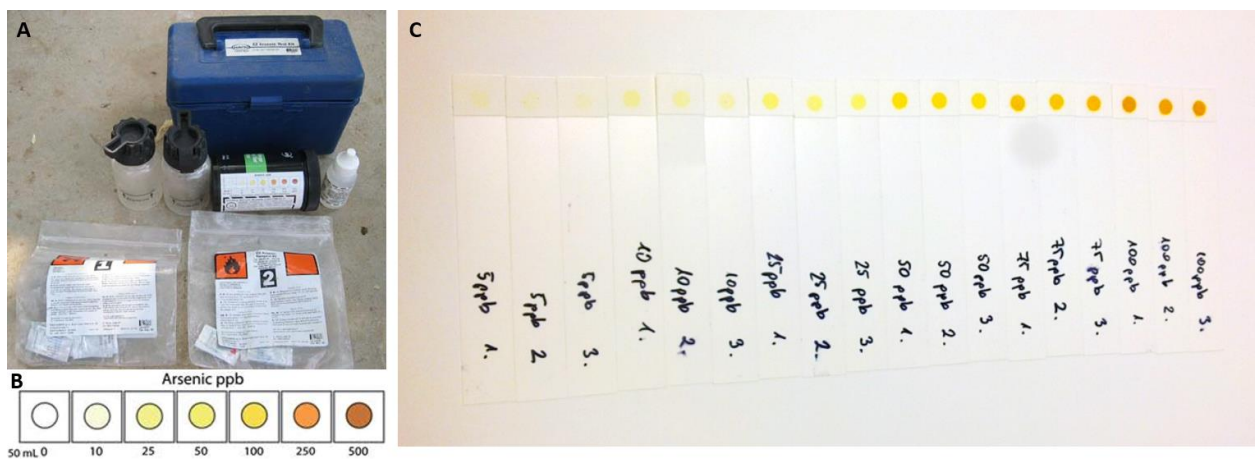

Fig. 1. (a) The Hach® EZ arsenic field test kit with its reagents; (b) the reference color series given by the manufacturer to compare the results to; (c) photograph of a series of experimental calibration results performed in triplicates by using the original reaction vessel and original reagents.

3. Results and Discussion

The characteristics of our integrated colorimetric sensor is presented in Fig. 4. As can be seen, our device has a linear response to arsenic concentrations in the 5-20 µg/l (ppb) range (Fig. 4), so is capable to distinguish between drinkable and not-drinkable water based on the World Health Organization (WHO) guidelines' limit of 10 µg/l. A slight drawback of this method is that it takes at least around 60 min to yield reliable results in the relevant concentration range at generally occurring ambient temperatures of drinking water (e.g. between 10-25 ºC).

To reduce the response time of the device, the effect of elevated water sample temperature was investigated. Sample temperatures of 25 ºC, 40 ºC, 60 ºC and 80 ºC were used and the appearance of the inflexion point of the kinetic curves was determined at a fixed arsenic concentration of 20 µg/l. Temperature values refer to sample temperature at the beginning of measurements, temperature control during the measurement is not integrated in the developed system.

At 80 °C, only 3 successful measurements were performed due to the frequent leakage of the microfluidic cartridge and the vapor formation due to high temperature. The effect of the elevated water sample temperature on the appearance of the inflexion point of the kinetic curves is shown in Fig. 7. The average time until the appearance of the inflexion point at 60 °C is less than 50 % of that at 25 °C. The appearance of the inflexion point is one parameter that could be used to approximate the concentration of arsenic in the sample, higher concentration implies earlier appearance. However, due to its high standard deviation, this parameter was only used for experimental purposes, not for arsenic measurement, and thus its evaluation was not implemented in the final construction of the device.

The appropriate sample temperature was determined based on the following considerations. Higher temperature enables faster approximation of concentration, however vapor formation becomes significant above 60 °C. Due to this, an overshoot appears on the kinetic curves, making the early approximation unreliable. Sample preparation and handling also requires more care at higher temperatures. Considering these effects, 50 °C was chosen for further measurements with different concentrations of arsenic in the sample. The time to the appearance of the inflexion point of the kinetic curves in function of the arsenic concentration of the sample (5, 10, 15, 20 and 50 μg/l, respectively) is shown in Fig. 6.

The kinetic curves and distribution of the relative blue component values after 25 min measurement time at 50 °C for the different arsenic concentrations are shown in Fig. 5. Measurements were performed in triplicates. Static characteristic plotted using these values is shown in Fig. 4. According to these results, concentration values can be reliably distinguished by the associated relative blue component values after only 25 min in this elevated temperature.
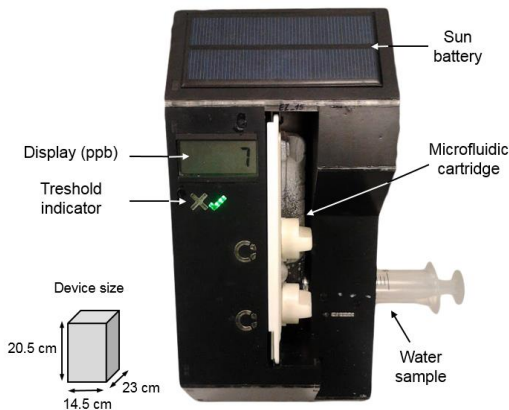

Fig. 2. The custom-developed colorimetric device for arsenic detection, which was used for the experiments.
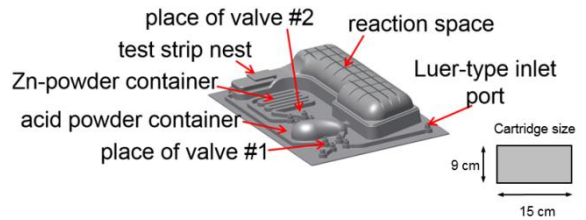

Fig. 3. 3D illustration of the custom microfluidic cartridge, including the reagents and the reaction chamber.
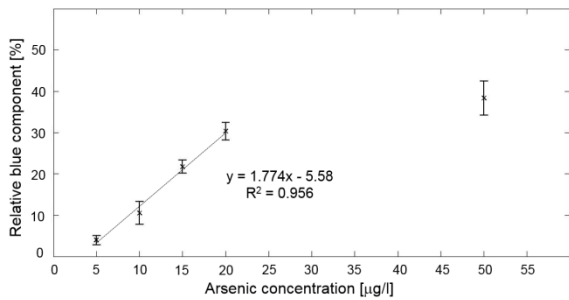

Fig. 4. Static characteristic of the colorimetric sensor: relative blue component in function of the arsenic concentration measured after 25 min at 50 °C. Average based on 5-13 measurements.
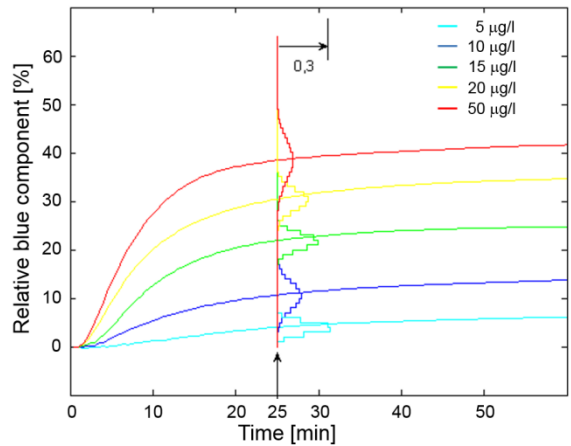
$$y = 1.774x - 5.58$$
$$R^2 = 0.956$$

Fig. 5. Kinetic curves of the measurements done at 50 °C with various arsenic concentrations. The superposed distributions at 25 min are calculated based on 5-13 measurements.
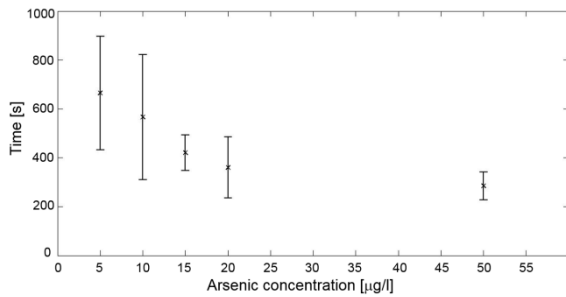
Fig. 6. Appearance of the inflexion point of the kinetic curves in function of the arsenic concentration of the sample, measured at 50 ºC. Average and standard deviation of 6-13 measurements.
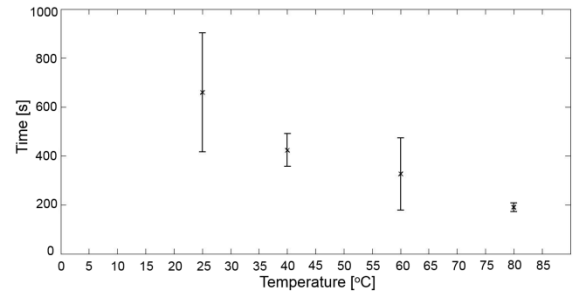

Fig. 7. Appearance of the inflexion point of the kinetic curves in function of the sample temperature, measured at 20 µg/l arsenic concentration. Average and standard deviation of 6-15 measurements, except at 80 ºC with only 3 measurements.

4. Conclusions

The capabilities of our custom-built colorimetric device were demonstrated to measure the arsenic concentration of drinking water samples. To decrease the measurement time the effect of elevated water sample temperatures was investigated in the range of 25-80 ºC. It was found that the time until the appearance of the inflexion point in the kinetic curves correlates with the arsenic concentration, however due to its high deviation it was not included in the evaluation process. It was demonstrated that at an elevated temperature of 50 ºC the arsenic concentration of the drinking water samples can reliably be obtained only after 25 min (compared to the original 60 min at ambient temperatures) and that our device has a linear response to arsenic concentrations in the 5-20 µg/l (ppb) range. Based on this result, our device is capable to determine the approximate arsenic concentration of water samples with calculable probability, and reliably distinguish drinkable water based on the 10 µg/l WHO guideline, as its primary purpose.

Acknowledgements

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