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Development of a Visual pH Measurement Method Using a Wedge-shaped Microspace with pH-dependent Swelling Hydrogel Microparticles and Its Application to Creatinine Measurement

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In this study, we developed an instrument-free visual pH determination method using a wedge-shaped microspace with pH-dependent swelling hydrogel particles. This method of pH measurement is achieved by utilizing the property of the hydrogel to swell with increasing pH and the wedge-shaped microspace in a 3D structure. Hydrogel particle formation using a microdroplet-forming chip and an ultraviolet LED enabled the fabrication of spherical pH-responsive hydrogel particles. Microchips with a wedge-shaped microspace of 500–3500 μm could be fabricated using the structures created by a photo-engineered 3D printer as templates. The evaluation of the method using a pH standard demonstrated that the settling distance of the particles decreased with increasing pH, and that the sensitivity and pH resolution were estimated to be 2.94 mm/pH and 0.2 pH, respectively. These results indicated that the method has the potential to visually measure pH. Furthermore, the developed method was applied to the determination of creatinine using creatinine deiminase. The method is readily applicable to biosensing technologies based on pH transduction, making it useful for on-site analysis in a wide range of fields, including medicine, environment, and food. Moreover, it can be adapted to microfluidic analysis for visual quantification using device-free micrototal analysis systems or lab-on-a-chip.

1. Introduction

pH is a quantitative measure of the acidity or basicity of aqueous or other liquid solutions, and is the negative logarithm of the hydrogen ion concentration in the solution. The measurement of pH is routinely performed in various fields such as healthcare,(1) environment,(2) food,(3) and

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agriculture.[4] Recently, the demand for pH measurement in the field has been increasing, especially in the field of medical testing, and the development of biomarker diagnostics[5–7] based on signal conversion to pH has been actively pursued. In the past, pH measurement methods using handheld pH meters or pH test strips and colorimetric pH measurement methods have been applied to biosensing as methods that enable pH to be measured easily in the field. pH meters[8,9] can provide high-resolution pH measurements. However, these measurements require specialized equipment for measuring potential, routine calibration and maintenance, and initial training for use. Therefore, when the quantification of target substances based on pH measurement is carried out in the field, an analysis method that can be easily used by anyone without requiring specialized equipment is desired. On the other hand, pH measurement methods based on "color change," such as the use of pH test papers[10] and the colorimetric quantification method,[11] are suitable for on-site measurements because they can determine pH without using equipment. However, pH test papers can discriminate approximate pH differences but have low pH resolution. Conversely, the colorimetric quantification method is regarded as a semiquantitative analysis, as it estimates pH by comparing the color presented by pH with a color sample. These color-change-based pH measurement methods are subject to the common problem that the estimated pH may differ depending on the visual color perception of the tester.

In more recent times, the pH measurement method that quantifies the intensity of the colors in images obtained with smartphones or tablet devices[12] has also been employed. This method enables the objective evaluation of differences in color tone, but the color intensity obtained may vary depending on the type of equipment and the environment in which the photographs are taken.[13] Furthermore, specialized software and operating skills are required for analysis. Therefore, there is a need for a simple and quantitative pH measurement method that can yield consistent results regardless of the location and the individual performing the measurement.

In this study, a pH visual measurement method using a wedge-shaped microspace with pH-dependent swelling hydrogel microparticles was developed. Previously, we have developed a method of measuring the size of microparticles using a wedge-shaped microspace.[14] By adapting this method to measure the size of pH-dependent swelling hydrogel particles, it was considered possible to visually measure pH. The distance of their settling can be measured visually, allowing for the quantitative and straightforward measurement of pH without the use of equipment. The measurement principle was demonstrated in experiments using pH-responsive hydrogel particles[15] consisting of acrylic acid (AA) and poly(ethylene glycol)-block-poly(propylene glycol)-block-poly(ethylene glycol) (Pluronic F-127),[16] in conjunction with buffer solutions exhibiting disparate pH values. Furthermore, the developed pH measurement method was also applied for the measurement of creatinine, which is one of the renal function markers,[17] and the results showed the potential of this method for biomarker measurement.

2. Materials and Methods

2.1 Reagents and materials

All reagents utilized in this study were of analytical reagent grade unless otherwise stated. The water used in experiments was purified by a PURELAB flex3 (Veolia Jenets K.K., Tokyo,

Japan). The 3D print resin for the fabrication of the microchip mold was obtained from Shenzhen Anycubic Technology Co., Ltd. (Shenzhen, China). The transparent resin for the fabrication of the microchip with a wedge-shaped microspace was obtained from Shoeitsusho Co., Ltd. (Aichi, Japan). The polydimethylsiloxane (PDMS) prepolymer and curing agent were purchased from Dow Corning Toray Co., Ltd. (Tokyo, Japan). Acrylic acid (AA) and lithium phenyl(2,4,6-trimethylbenzoyl) phosphinate (LAP) were procured from Tokyo Chemical Industry Co., Ltd. (Tokyo, Japan). The poly(ethylene glycol)-block-poly(propylene glycol)-block-poly(ethylene glycol) dimethacrylate (Pluronic F-127 DMA) and platinum(0) 1,3-diethenyl-1,1,3,3-tetramethyldisiloxane complexes (Karstedt's catalyst) were obtained from Sigma-Aldrich (MO, U.S.A.). Mineral oil was purchased from Nacalai Tesque, Inc. (Kyoto, Japan). Standard solutions for pH measurement, glucose, isopropyl alcohol (IPA), and ethanol were obtained from Kanto Chemical Co., Inc. (Tokyo, Japan). Brilliant blue FCF (blue dye), ammonia, creatinine, and LabAssay^{TM} Ammonia were procured from Fujifilm Wako Pure Chemical Corp. (Osaka, Japan). Creatinine deiminase (CD) was obtained from MP Biomedicals, Inc. (CA, U.S.A.).

2.2 Preparation of pH-responsive hydrogel microparticles

A microchip template for droplet generation was designed using 3D CAD software (Fusion 360, Autodesk Inc.). The stereolithography data were processed to create slice images using a 3D printer and special slicer software (Photon Workshop V2.1.30.RC17). The microchip template, comprising a structure formed from colored UV resin, was created using a 3D printer (Anycubic Photon M5, Anycubic Inc.). The slice data was loaded into the printer, which then produced the printed structure. This structure was immersed in IPA for 10 min and then rinsed with IPA until the unreacted resin was removed. The structure was then dried at room temperature (RT), and a gold thin film (approximately 10 nm) was formed on the top surface of the structure using a magnetron sputtering system. The PDMS prepolymer (containing a very small amount of catalyst) and curing agent were mixed at a weight ratio of 10:1, and this mixture was poured into the microchip template. The mixture was then cured at RT overnight. The PDMS plate replicating the channel was peeled from the template, and holes (1.5 mm in diameter) were then formed using a biopsy punch (BP-15F, Kai Industries Co., Ltd.). Figure 1(a) shows the structure of the PDMS microchip for microdroplet generation. The width and depth of the microchannels ranged from 500 to 1000 μm.

A reaction solution containing 62% (w/w) AA, 4.6% (w/w) Pluronic F-127 DMA, 1.0% (w/w) LAP, and 0.10 M blue dye was pumped at 1.0 μL/min from Inlet 1, and mineral oil was pumped at 6.0 μL/min from Inlet 2, to form water-in-oil microdroplets in the channel. Subsequently, the droplets were directed towards the collection reservoir, where they were subjected to 405 nm light emitted from an LED (405 nm, OSSV5131A, OptoSupply Ltd., N. T., Hong Kong). This light triggered photopolymerization, resulting in the formation of spherical hydrogels [Fig. 1(b)]. Subsequently, the hydrogel particles were transported to the collection chamber and washed with phosphate buffer (PB) (pH 7.40) to eliminate any unreacted monomer, cross-linker agents, and blue dye. They were then immersed with fresh PBS for storage before use.

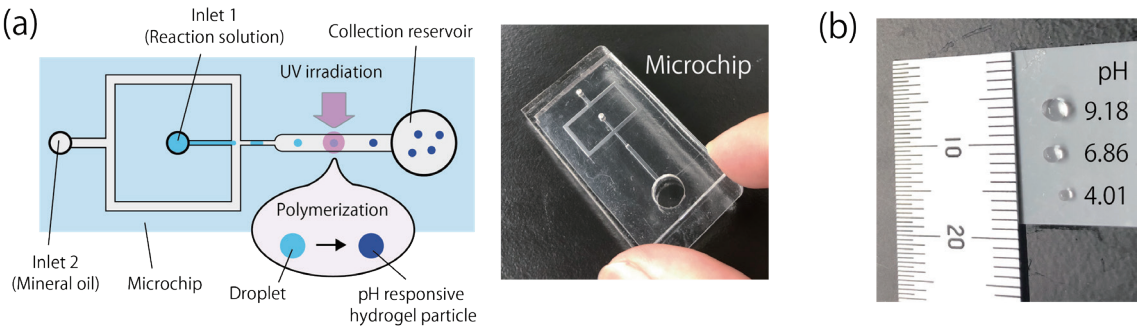


Fig. 1. (Color online) (a) Illustration and photograph of the microfluidic device for microdroplet formation. (b) Photograph of the prepared pH-responsive hydrogels.

2.3 Fabrication of microchip with a wedge-shaped microspace

The microchip with a wedge-shaped microspace was fabricated using a 3D printer. The design and photograph of the fabricated microchip are shown in Fig. 2. The microchip is 55 mm in length, 20 mm in width, and 5 mm in height. The wedge-shaped inclined space within the chip measures 50 mm in length and 15 mm in width, with a maximum height of 3500 μm and a minimum height of 500 μm. The volume of the space is 1.5 mL. Mineral oil and hydrogel particles were introduced into the microchip through an inlet at the top during the pH measurement.

2.4 Visual pH measurement using pH standard solutions

The principle of the visual pH measurement is shown in Fig. 3. pH-responsive hydrogel particles were dried in an oven at 70 °C for 10 min before use. Subsequently, the particles were immersed in the sample solution for 30 min. After a specified period, the particles collected from the sample solution were introduced into the inlet of a wedge-shaped microspace filled with mineral oil. The particles settle in the wedge-shaped microspace and come to a stop when they collide with the wall. The distance from the inlet to the position of the center of the stopped particles was measured, and pH was estimated on the basis of this distance.

2.5 Creatinine measurement

The principle of the creatinine assay is shown in Fig. 4. A creatinine solution (0–100 mg/dL) in 5.0 mM PB (pH 7.40) was prepared and mixed with 5.0 U/mL CD solution in a 1:1 volume ratio in a microtube. To allow the reaction to proceed, the microtubes were incubated at room temperature for 60 min. Subsequently, the dried pH-responsive hydrogel particles were immersed in the sample for 10 min, after which they were introduced into a wedge-shaped inclined space, and the settling distance was then measured.

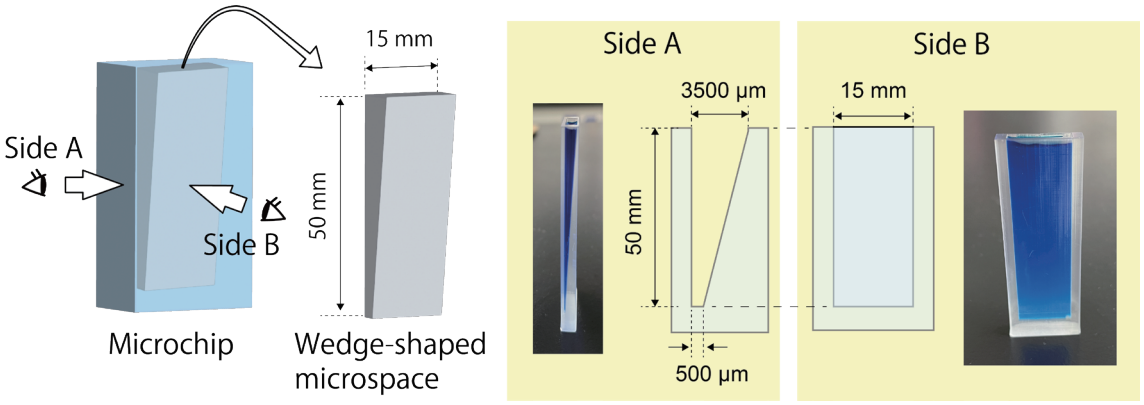


Fig. 2. (Color online) Design and photograph of a microchip with a wedge-shaped microspace. In the picture, the microspace is filled with a blue dye solution.

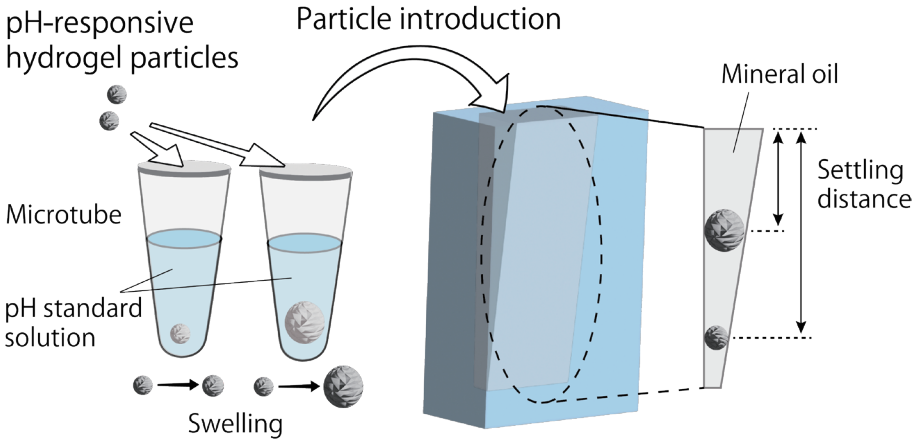


Fig. 3. (Color online) Schematic illustration of the principle of pH measurement using pH-responsive hydrogels and a wedge-shaped microspace.

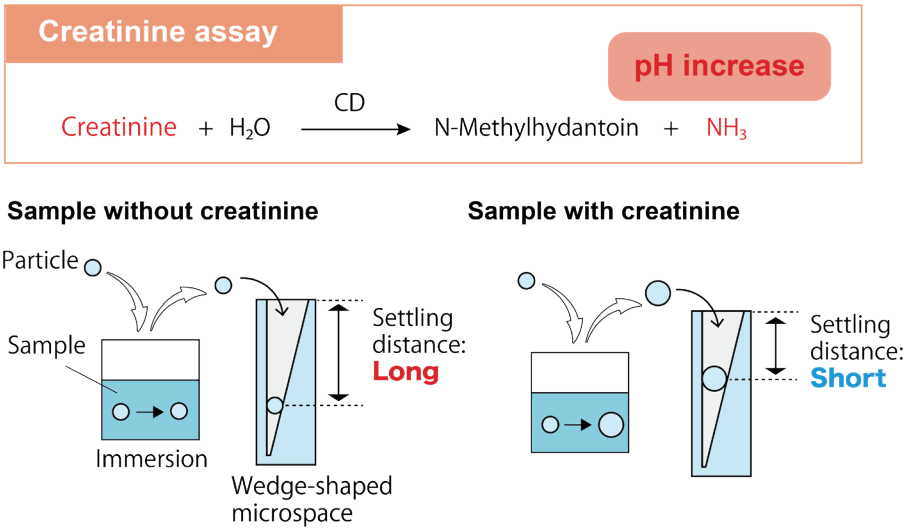


Fig. 4. (Color online) Schematic illustration of the principle of creatinine assay.

3. Results and Discussion

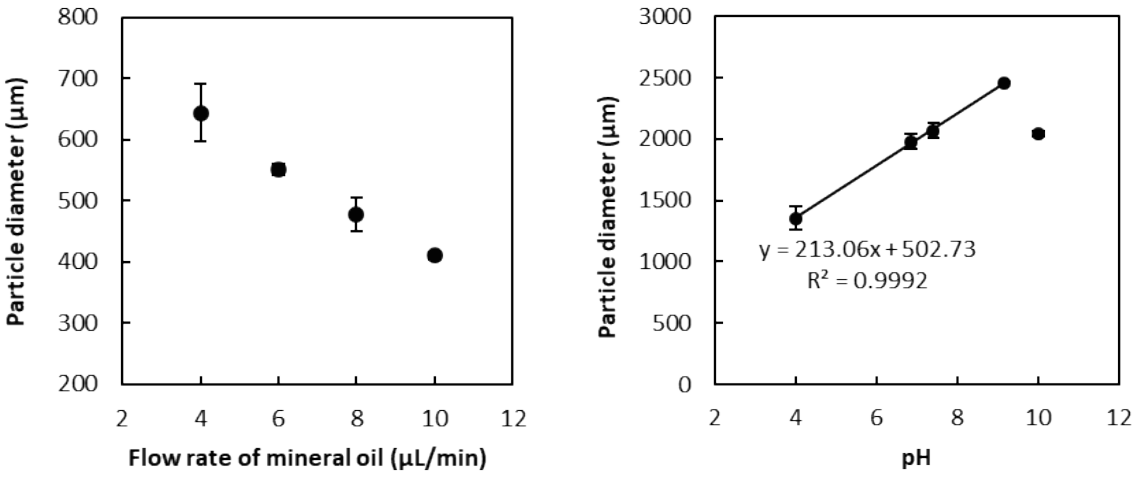
3.1 PDMS microfluidic device for droplet generation

A structure used as a template for the microfluidic device for droplet generation was fabricated by printing on a 3D printer under default conditions. A mixture solution of the PDMS main agent and the curing agent at a weight ratio of 10:1 was poured into the mold with the template and heated at 70 °C for 1 h. However, the PDMS in contact with the mold did not cure effectively. The reason for this is assumed to be that the components in the 3D-printed structure interfere with the PDMS cross-linking reaction. To address this issue, a gold thin film coating on the surface of the structure was investigated through gold sputtering[18] and the addition of a PDMS catalyst containing platinum. The results demonstrated that the microstructure could be successfully transferred to PDMS when the thickness of the gold thin film was approximately 10 nm and the catalyst addition was 0.10% (w/w).

3.2 pH-responsive hydrogel microparticles

The droplet generation method using microchannels can be employed to form microsized droplets with a high degree of reproducibility, with a small variation in particle size.[19] Hydrogel microparticles can be formed by flowing the reaction solution containing monomers, cross-linking agents, and polymerization initiators, and mineral oil into a cross-shaped microchannel to form water-in-oil microdroplets, which are then irradiated with light to promote a polymerization reaction.[20] Here, the conditions to produce pH-responsive hydrogel microparticles were investigated using a self-made droplet-forming microchip. The flow rate of the reaction solution was maintained at a constant level, and the ability to form particles was evaluated when the oil flow rate was varied. The relationship between the oil flow rate and the hydrogel particle size, as well as the images of the hydrogel particles obtained, are shown in Fig. 5(a). The diameter range of the particles produced was approximately 400–700 μm, with particle size decreasing with increasing oil flow rate. The variation in the size of the particles produced under all conditions was less than 7.3%. This indicates that pH-responsive hydrogel particles can be stably prepared by this method. An optimal oil flow rate of 6 μL/min was selected to produce particles with a visually observable size and the lowest size in variation.

When the prepared dried particles were placed in pH standard solutions (pH 4.01, 6.86, 7.41, 9.18, and 10.01) for 30 min, all particles swelled [Fig. 5(b)]. This is attributed to the swelling of the hydrogel particles as the solution permeates into the dried hydrogel particles. The size of the hydrogel particles exhibited a linear increase with pH, with a change of 213 μm per pH (linear approximation: $y = 213x + 503$). This pH-dependent swelling of the gel is caused by the repulsion of the charges of the carboxylate ions present inside the gel. It is assumed that the higher the pH of the solution, the more carboxylate ions are formed in the gel and the particles swell owing to their repulsion. This result suggests that pH-responsive hydrogels can be produced by the present method. On the other hand, when a standard solution of pH 10.01 (carbonate buffer) was used,



(a) (b)

Fig. 5. Evaluation of pH-responsive hydrogel microparticles. (a) Relationship between oil flow rate and hydrogel particle size. Unwashed particles, formed in the microfluidic channel and collected immediately, were used for the measurements. Error bars indicate the standard deviations from three trials. (b) Changes in hydrogel particle size when immersed in different pH buffers. Error bars indicate the standard deviations from three trials.

the particle size was smaller than when that of pH 9.18 (borate buffer) was used. This is considered to be due to the high content of sodium ions in the carbonate buffer solution, resulting in a higher ionic strength and a lower electrostatic repulsive force between carboxylate ions. Experimental results demonstrated that the particle size of the gel exhibited a roughly linear change between pH 4.01 and 9.18, with a change of approximately 1400 to 2500 μm. Therefore, a wedge-shaped microspace with a maximum height of 3500 μm and a minimum height of 500 μm was designed to be used for pH measurement.

3.3 Demonstration of pH measurement

Hydrogel particles immersed for 30 min in buffer solutions at different pH values were introduced into a wedge-shaped microspace filled with mineral oil, and the distances at which they settled were measured. The relationship between the pH of the solution and the settling distance is shown in Fig. 6. The settling distance decreased as the pH of the solution increased. The settling distance increased when a buffer solution of pH 10.01 was used. These results correlated with the response of the particle size to the solution pH as shown in Fig. 5.

A calibration curve prepared in the pH range from 4.01 to 9.18 exhibited good linearity ($R^2 = 0.9969$), with a linear approximation of $y = -2.94x + 47.9$. The sensitivity was estimated to be 2.94 mm/pH from the slope of the calibration curve, and the pH resolution was calculated to be 0.2 pH (since the minimum length measurable by visual observation is approximately 0.5 mm, the pH resolution was defined as the pH value corresponding to 0.5 mm). This result indicates that pH can be measured visually by this method. The method has the potential to achieve a higher pH resolution through the expansion of the wedge-shaped microspace.

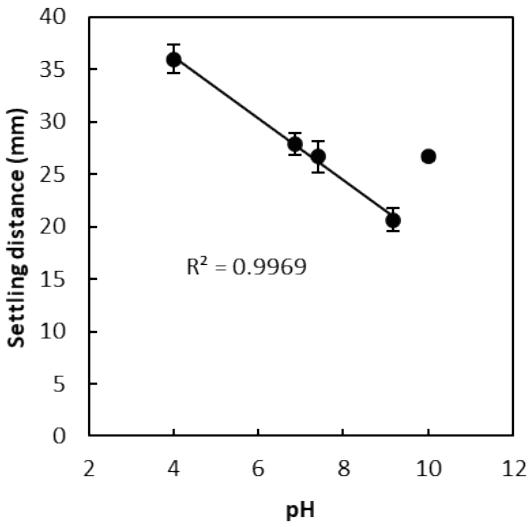


Fig. 6. Relationship between settling distance and pH of the solution. Error bars indicate the standard deviations from three trials.

A comparison table between this method and other pH measurement methods is shown in Table 1. The pH resolution of this method is lower than that of the electrochemical method using a pH electrode. However, the developed method requires no equipment, enables visual pH measurement, and offers a pH resolution higher than that in the case of using pH test papers. Furthermore, this method has the potential to achieve an improved pH resolution by expanding the wedge-shaped microspace, making it a promising tool for a simple and accurate pH measurement in the field.

3.4 Creatinine assay

Creatinine is one of the biomarkers of renal function in human serum.[17] In the enzyme reaction occurring in the presence of CD, the pH of the solution increases owing to the production of ammonia from creatinine. First, to investigate the relationship between ammonia concentration and pH, the pH of 0–5.0 mM ammonia solutions prepared with PB was measured. As shown in Fig. 7(a), pH increased with ammonia concentration and showed a linear correlation at ammonia concentrations of 0–1.25 mM. Next, creatinine solutions (0–100 mg/dL) were then mixed with 5.0 U/mL CD solution and the concentration of ammonia in the solution after the reaction was measured. The results showed that the concentration of ammonia corresponding to a creatinine concentration of 0–100 mg/dL was around 0–4.0 mM [Fig. 7(b)]. This result indicates that ammonia is produced by the reaction in the presence of creatinine deiminase and that the amount of ammonia in solution increases with creatinine concentration. Considering the results shown in Fig. 7(a), there was a possibility of a linear correlation between creatinine concentration and pH when creatinine solutions below roughly 25 mg/dL were used. Actually, the measurement of the pH of the solution after the reaction showed a linear correlation between creatinine concentration and pH in the concentration range of 0–12.5 mg/dL [Fig. 7(c)]. On the other hand, pH did not increase significantly at concentrations above 25 mg/dL and was lower than that measured for the ammonia solution [Fig. 7(a)]. This might be due to the presence of CD

Table 1
Comparison of this method with other methods.[21]

Method	Notes	Resolution (pH units)	Advantages and disadvantages
Potentiometric method	Electrochemical measurement normally made using pH probe	0.05	• High resolution • Handling, care, and maintenance of probe
Photometric method	Spectrophotometer to measure wavelength of pH-sensitive colored solution	0.1	• Higher resolution than visual method • Requires solution to be colorless
Visual method with pH paper	Color comparison with pH-sensitive indicator paper or solution	0.5	• Quick and easy • Low resolution • Relies on human eye
Present method	Measurement of particle size of hydrogels, whose volume changes depending on pH of solution, with wedge-shaped microspace.	0.2	• Quick and easy • High resolution regardless of visual observation • Further increase in resolution by designing hydrogel and microspace

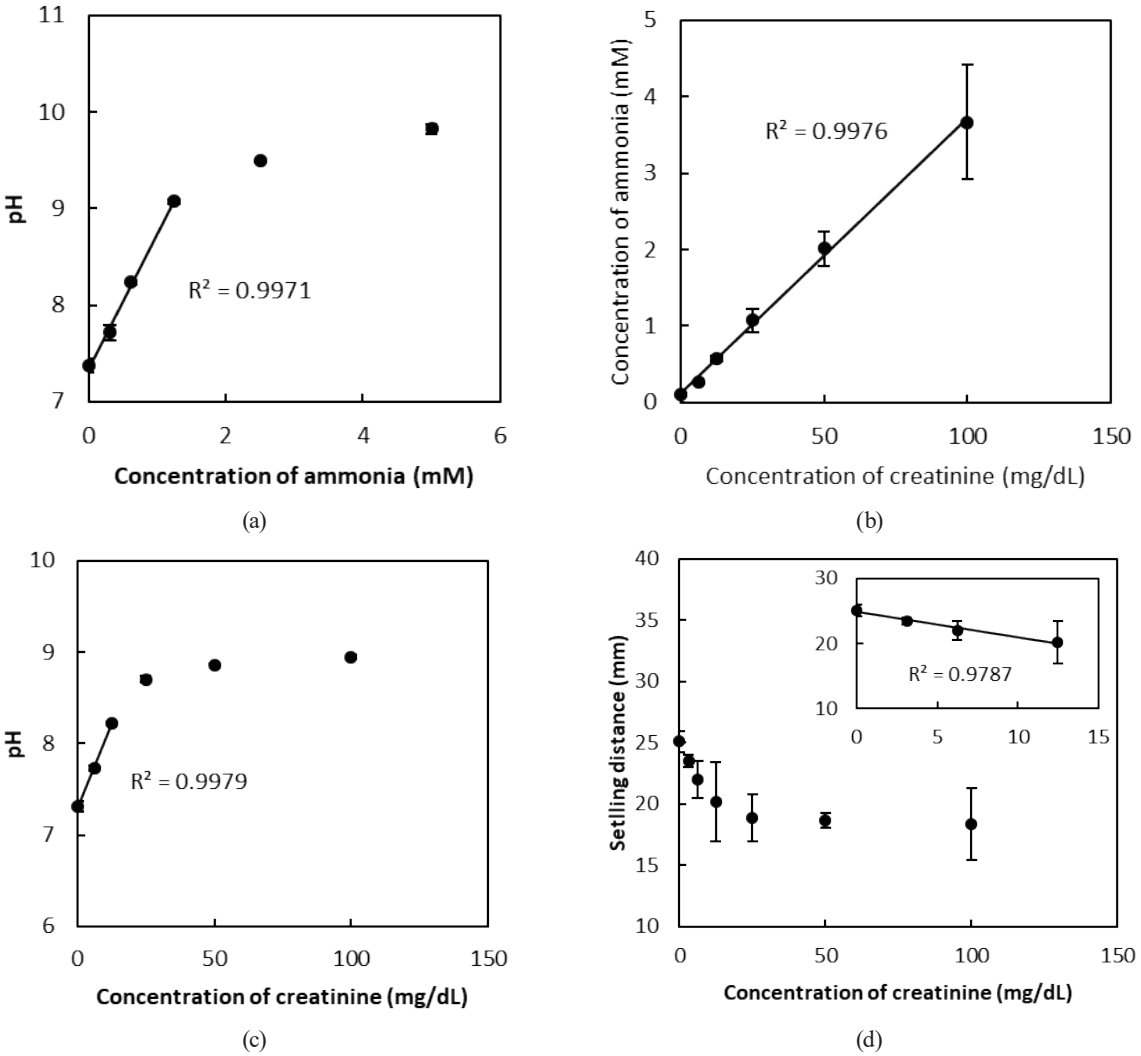


Fig. 7. Experimental results for the measurement of creatinine. (a) pH against concentration of ammonia. (b) Concentration of ammonia in solution after reaction against creatinine concentration. (c) pH of the solution after the reaction against creatinine concentration. (d) Relationship between creatinine concentration and settling distance. The small graph in the top right-hand corner shows calibration curves prepared for a concentration range of 0–12.5 mg/dL. Error bars indicate the standard deviations from three trials.

and *N*-methylhydantoin produced in the reaction. The developed method was used to measure 0–100 mg/dL creatinine solutions. Following the enzyme reaction, the pH-responsive hydrogel particles were immersed in the solution, and the settling distance was measured in a wedge-shaped microspace. The results obtained are shown in Fig. 7(d). The settling distance decreased with increasing creatinine concentration and remained almost constant at creatinine concentrations above 25 mg/dL. The change in sedimentation distance correlated well with the change in pH shown in Fig. 7(c).

The calibration curve for creatinine prepared in the concentration range of 0–12.5 mg/dL showed linearity ($R^2 = 0.9787$), and the relative standard deviation (*RSD*, $n = 3$) obtained in that concentration range was estimated to be ≤16%. *RSD* increased with creatinine concentration. This is due to the greater swelling of the hydrogel particles with increasing creatinine concentration. Large hydrogel particles absorb more water than small hydrogel particles and are therefore softer and more likely to stop in different positions. In addition, when the particles swell to a nonuniform shape (i.e., ellipsoid), hydrogel particles with large diameters are more prone to errors in settling distance owing to their greater change in shape. These effects may be reduced by changing the concentration of monomers and cross-linkers in the reaction solution used in hydrogel preparation and by improving the exposure conditions during the polymerization reaction. Moreover, it might be possible to control the amount of change in particle size relative to the analyte concentration by selecting the pH and salt concentration of the buffer solution. Although the developed method requires further improvements in analytical performance, the experiments demonstrated the potential of this method for the measurement of biomarkers.

4. Conclusions

A pH measurement method using a wedge-shaped microspace with pH-dependent swelling hydrogel particles has been developed. The principal advantage of this method is that the pH of the solution can be determined by visually observing the settling distance of the pH-responsive hydrogel particles, obviating the need for instruments. The measurement principle was demonstrated using pH buffer solutions, and the method was successfully applied to the measurement of creatinine. The obtained results indicate the potential applicability of this measurement method to biosensors. The pH resolution of the method can be enhanced by optimizing the concentration of monomers and cross-linkers in the droplets and reducing the variation in the size of the pH-responsive hydrogel particles. Additionally, the pH resolution can also be improved by increasing the length of the wedge-shaped microspace. These considerations could be developed into a technique for visually measuring pH with high sensitivity. The method can be applied to the pH measurement of biological samples such as urine and sweat, as well as to biosensing based on pH transduction using urease or cholinesterase. Furthermore, the method can be easily applied to micrototal analysis systems and lab-on-a-chip, and is expected to contribute to the development of on-site analysis in a wide range of fields such as medicine, environment, and food.

Acknowledgments

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