



A Wireless Chemical Sensing Scheme using Ultrasonic Imaging of Silica-Particle-Embedded Hydrogels (Silicagel)



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ABSTRACT

In this paper, we introduce a wireless chemical sensing technique featuring ultrasonic imaging of a silica-bead-embedded hydrogel, named “silicagel”. By incorporating silica beads within a hydrogel network, its volume change in response to environmental stimuli can be remotely interrogated by ultrasonic imaging, either by directly measuring its dimensions (cross-section of the silicagel) or indirectly evaluating the back-scattered wave intensity. Various sizes (4 and 2 mm cube) and concentrations (0.05 and 0.1 w/v%) of silicagel fabricated with pH-sensitive poly (methacrylic acid-co-acrylamide) hydrogel is studied *in-vitro*, using a 40 MHz ultrasonic imaging system. For the silicagel with 4 mm cube and 0.05 w/v% (as a representative), its cross-section sensing method results in a sensitivity of 0.83 (mm²/mm²)/pH with a theoretical resolution of 0.001 pH while back-scattered intensity technique exhibit a sensitivity of 17.5 gray-scale intensity/pH with a resolution of 0.06 pH unit in the maximal response region (between pH 4 and 6). It is anticipated that the same technique can be applied to hydrogels sensitive to other stimuli (e.g., glucose, specific ions, biomarkers, etc.).

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1. Introduction

In-vivo wireless electrochemical sensing techniques have been extensively investigated since the ingestible “radio pill” first demonstrated by MacKay in 1957 [1]. Most such techniques demand an on-board power source to activate the electrochemical sensing element. More recently, however, the chemo-mechanical transduction of hydrogels, water-absorbable polymers, have been employed to fabricate passive wireless chemical sensors eliminating the need for the on-board power source [2–14]. Environmentally sensitive-hydrogels are able to transduce a chemical signal (e.g., pH or blood glucose level) to a mechanical one (e.g., pressure or volume change), enabling *in-vivo* battery-less operation [15–27] (before their application to sensing, the volume and shape responses of hydrogels were utilized in autonomous actuators and

drug delivery systems [28–36]). Our group first reported a MEMS-based passive LC transponder, whose capacitance was dependent on osmotic swelling and shrinking of a pH/glucose responsive hydrogel [37]. However, the complicated fabrication process associated with a hermetically sealed MEMS capacitor led to a low yield and high manufacturing costs.

Alternative hydrogel-based sensing and actuation mechanisms based on the incorporation of micro/nano scale materials onto the polymeric network of hydrogels have also been reported by several groups [38–47]. For instance, a chemical sensor based on a crystalline colloidal particles polymerized within a hydrogel allowing its color to change in response to glucose was reported by Holtz et al. [46]. Another effort by our group integrated magnetic nanoparticles with a pH-sensitive hydrogel to develop a low-cost and easy-to-fabricate passive LC resonator [47]. The magnetically functionalized hydrogel, termed “ferrogel,” which was prepared by physically embedding the nanoparticles inside of a pH sensitive hydrogel, possesses volume-controlled magnetic permeability (i.e., lower permeability in the shrunken state and higher permeability at the swollen state) that can be used in conjunction with a planar coil to form a passive LC transponder whose inductance is modulated with pH variations. While the fabrication process of the ferrogel sensor was significantly simpler than the MEMS-based

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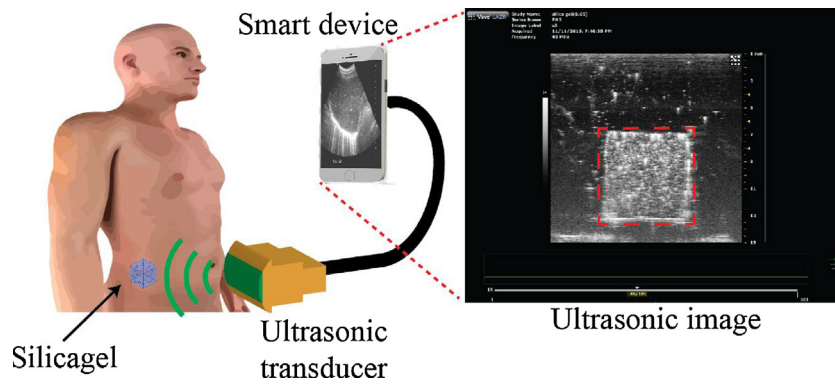


Fig. 1. Schematic of an envisioned application for the silicagel. The silicagel is implanted either subcutaneously or deep within the body cavity and is imaged ultrasonically. The volume transition due to chemical stimuli can be detected by directly measuring the hydrogel cross-sectional area or the intensity of the back-scattered wave.

one, the short interrogation range (~1 cm) and the requirement for a complicated readout circuitry limited their practical applications to scenarios in which the sensor is implanted subcutaneously.

In this paper, we demonstrate a different chemical sensing method featuring a silica-bead-embedded hydrogel (silicagel) whose volumetric response to the chemical stimuli can be interrogated by ultrasonic imaging. Since ultrasonic waves at clinical imaging frequencies (2–20 MHz) is capable of deep tissue penetration (>10 cm) [48], the described method removes one of the major bottlenecks of passive sensing. In addition, this technique can be clinically relevant since ultrasonic imaging equipment is commonly available in most clinics.

2. Working principle

Fig. 1 shows a schematic illustration of the sensor and its envisioned application. The functionalized hydrogel, in which silica beads are physically embedded during polymerization, designated “silicagel,” can be inserted in either the subcutaneous space or deep tissue within the body. The volume transition of environmentally sensitive hydrogel alters the density of the silica beads, leading to a change in intensity of back-scattered ultrasonic wave and its transformation into an ultrasonic image. This enables a noninvasive biochemical monitoring scheme that only requires a minimally invasive surgical procedure for implantation of the silicagel. Although the embedded silica beads have been extensively utilized as a carrier in drug delivery systems [49–53], this work is the first to report their use in conjunction with hydrogel for chemical sensing.

Ultrasonic imaging is basically enabled by the dissimilarity in acoustic impedances of two media, determining the reflection degree of the propagating acoustic wave. The reflection coefficient can be defined as

$$\Gamma = \frac{(Z_1 - Z_2)^2}{(Z_1 + Z_2)^2} \quad (1)$$

where Z_1 and Z_2 are acoustic impedances of the media; higher reflection coefficient, enhanced visibility to the ultrasound. For a bare hydrogel immersed in water, the difference in the acoustic impedance between hydrogel and water is very small, resulting in the diminished contrast in the ultrasonic image. The silica beads enhance the contrast of the hydrogel immersed in aqueous media. The dimension of the silica beads must be chosen such that the incoming ultrasonic wave would scatter back to the transducer from individual beads (i.e., beads must be smaller than the wavelength of the sound); in this paper, 9–13 μm diameter beads and 40 MHz transducer (~40 μm wavelength in water) were employed [54].

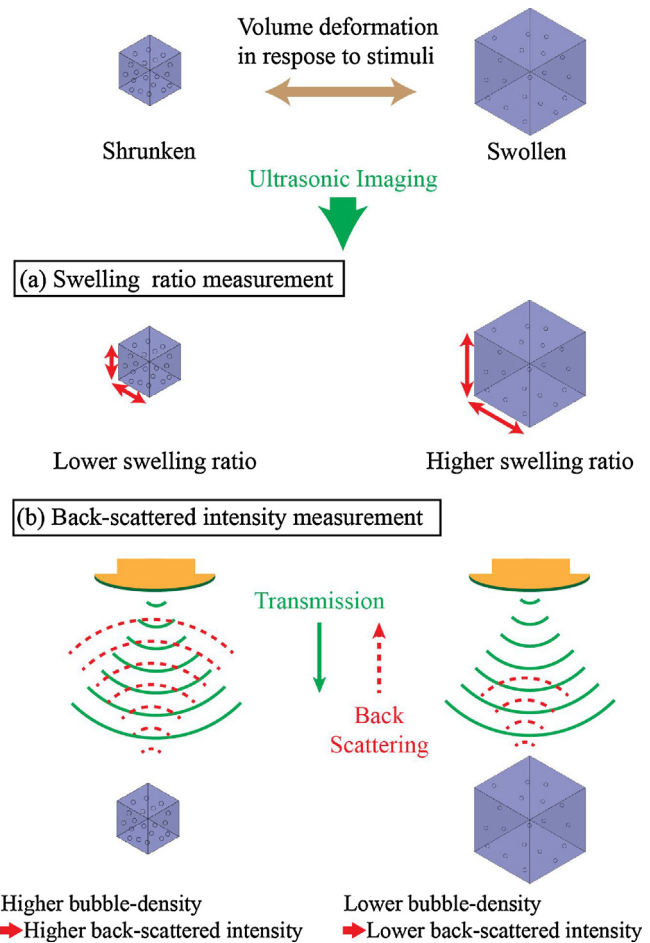


Fig. 2. Two sensing mechanisms of the silicagel sensor using ultrasonic imaging: (a) swelling ratio measurement and (b) back-scattered intensity measurement.

The sensing of chemical stimuli using the aforementioned property of silicagel is achieved by two different methods, as shown in Fig. 2. First, the increased contrast of the silicagel allows the direct cross-section measurement of silicagel from its ultrasonic image. Second, since the silica bead density of randomly selected region is modulated with swelling/shrinking of the hydrogel in response to the chemical stimuli (a higher density at in the shrunken state, and a lower density at in the swollen state); the ultrasonic image of silicagel can remotely detect the changes of the bead-density as a function of volume change by obtaining the back-scattered inten-

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