High-power Low-Q RF ESR Resonator for Pulse Techniques

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A new type RF ESR resonator for pulse techniques was developed. The resonator is an LC circuit that consists of a single turn coil (inner diameter, 45 mm) and ceramic capacitors. A two-turn coil-shaped polyethylene tube is located inside of the loop and an aqueous sodium chloride solution is perfused into the tube at a rate of 6 cm^3/min. Its resonant frequency is approximately 300 MHz, with a Q of less than 70. Using this resonator, pulsed-longitudinally detected ESR measurements of a phantom (inner diameter, 20 mm) containing a triaryl methyl spin probe were performed. The Q value was sufficiently low for irradiation over wide frequency band in the pulsed radiowave where the pulse width was 200 ns. The apparatus can handle a sufficient high power (maximum, 100 W) adjusted according to the sample size.

In vivo ESR spectrometers operating in the UHF band, where the dielectric loss is sufficiently small, have been developed to measure free radicals in lossy samples such as small animals (e.g., rats and mice).[1-7] In general, the continuous wave method has been used for ESR measurements because the relaxation times of the resonance system is short. So pulse techniques have been barely employed in this field. For irradiation over a wide frequency band in the pulse, it is necessary that the Q of the resonator is sufficiently low. When a resonant frequency becomes lower under the same pulse width, one must further reduce the Q of the resonator. By employing lossy elements in the resonator system, its Q will become lower. However, irradiated power is converted mainly into heat in the lossy elements; so it is difficult to use high power because the lossy elements will burn. Yet in the *in vivo* study, the power must be increased as the sample size increases. So far, one has not been able to utilize any methods to reduce both the Q and the heat in the resonator system. A low Q resonator measuring 6 mm in inner diameter for pulsed RF ESR was reported,[8] but it is so small that only a limited sample, such as the tail of a rodent, can be inserted. To overcome these problems, a new high-power low-Q resonator operating at 300 MHz was developed in this study.

This resonator is an LC circuit consisting of a single turn coil and ceramic capacitors. The former is a copper loop measuring 45 mm in inner diameter and 10 mm in axial length. Total capacitance of the latter is 5 pF. Under these conditions, the resonant frequency is 280 MHz and the Q, 570. A two-turn coil-shaped polyethylene tube (inner diameter, 0.75 mm; outer diameter, 1.5 mm) was placed inside the loop (Figure 1). With these parameters, the resonator Q remained almost constant. When water was added to the tube, the Q changed little. However, when an aqueous 0.9% (w/w) sodium chloride solution was added to the tube, the Q dropped drastically to 70. Here, the resonant frequency was 286 MHz. Bandwidth where half of applied power is absorbed in the resonator system with Q=70 at a resonant frequency of 300 MHz is 4.3 MHz. When full-width at half-maximum (FWHM) of a power spectrum of a pulse is 4.3 MHz at a center frequency of 300 MHz, the pulse width is 200 ns. Thus, the Q=70 resonator can cover a frequency band that is included in the pulse measuring 200 ns in width at a resonant frequency of 300 MHz. An aqueous sodium chloride solution is perfused into the polyethylene tube at a rate of 6 cm^3/min. Subsequently, pulsed radiowave (resonant frequency, 286 MHz; pulse width, 200 ns; repeat time, 2000 ns) is irradiated to the resonator at an average power of 10 W (maximum power, 100 W). Irradiation for 10 min did not cause any damage to the tube in the resonator. This means that the heat that is generated in the resonator system is efficiently dissipated when the solution in the tube is perfused.

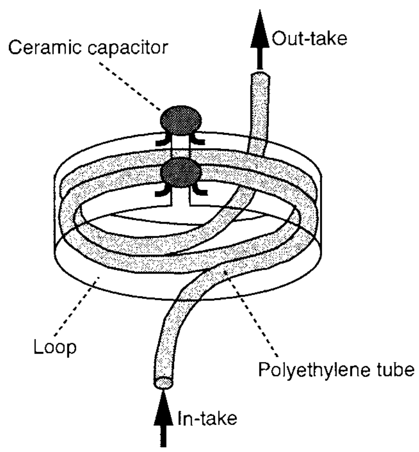

Figure 1. A scheme for our low Q resonator.

The Q values of the resonator were also observed for different turn numbers of the coil-shaped tube containing an aqueous sodium chloride solution. The respective Q of the resonator equipped with each tube having 2, 3, or 5 turns was 70, 44, or 30, showing that the Q can be tuned by changing the number of turns of the tube. The sample space of this resonator is 41 mm in inner diameter, which is adequate for the head of a rat or the whole body of a mouse.

Using this resonator, longitudinally detected ESR (LOD-ESR) measurements were performed. With an LODESR technique, the signal is derived from a longitudinal (i.e., parallel to the z-axis) oscillation of the magnetization caused by spin flipping under on/off modulated ESR irradiation.[9-13] We investigated changes in the shape of the LODESR spectrum under pulsed irradiation to see if the resonator in this study can successfully irradiate pulsed-radiowave.

The details of our LODESR spectrometer have been previously described.[11-13] A pair of saddle-type pickup coils

(STPCs) is used to detect the change in magnetization in the sample. Each coil of the pair is constructed from 15 turns of copper wire (0.3 mm in diameter). The outer diameter of the coil measures 30 mm. These coils are attached on a cylindrical quartz glass tube (38 mm in outer diameter). The resonator is located outside the STPCs. When a sample was irradiated by modulated radiowave power under the ESR conditions, the longitudinal change of the sample magnetization induced a signal in the pair of STPCs at a modulation frequency of 500 kHz. Conventional LC matching circuits were used to match the impedance of the pickup coils to the input impedance of preamplifiers at the modulation frequency. The Q of the STPCs is about 10. For the signal detection, the signals from the STPCs were pre-amplified, followed by the lock-in amplification at 500 kHz. The coil-shaped tube in the resonator has 2 turns. The aqueous sodium chloride solution was perfused continuously. The unloaded Q of the resonator was 51 when it was mounted on the STPCs and set in a shielded case for LODESR measurements.

The triaryl methyl spin probe, TAM, was a gift from Nycomed Innovation.[14] This water-soluble spin probe was stable in the solution throughout the experiments. Ten ml of a 1 mmol dm^{-3} solution of TAM that had been dissolved in a physiological saline solution (0.9 % sodium chloride aqueous solution) was placed in a sample tube (20 mm in inner diameter, 30 mm in outer diameter, 32 mm in axial length) to act as a phantom with a high dielectric loss (as in an *in vivo* sample).[12] When the phantom was inserted in the center of the resonator, the loaded Q of the resonator was 49 and the resonant frequency was 283 MHz. A pulse width of 200 ns is sufficient narrow because the relaxation time of the TAM aqueous solution is 670 ns.[15]

LODESR measurements of the phantom were performed at a pulse width of 200 ns and repeat time of 2000 ns (i.e., a modulation frequency of 500 kHz). By increasing irradiation power, the LODESR signal intensity of TAM reached a plateau

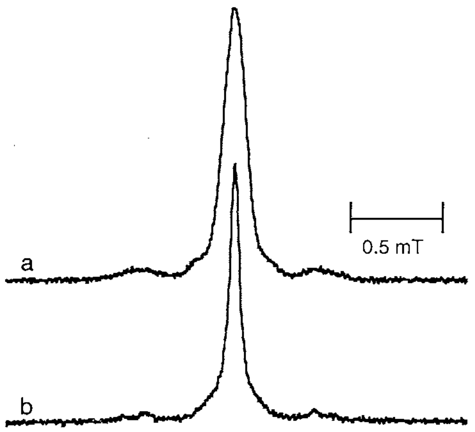

Figure 2. Spectra of a phantom obtained from pulsed-LODESR (a) and conventional LODESR (b). The phantom is a sample tube (20 mm in inner diameter, 30 mm in outer diameter, 32 mm in axial length) that contained 10 ml of a 1 mmol dm^{-3} solution of TAM dissolved in a physiological saline. The FWHM of the spectrum is 0.14 mT (a) or 0.06 mT (b). The instrument settings are: RF average power, 10 W; resonant frequency, 283 MHz; scan rate, 5 mT/s; accumulation number, 16; modulation frequency, 500 kHz; pulse width, 200 ns (a) or 1000 ns (b); time constant, 1 ms.

at an average power of 10 W (maximum power, 100 W). This suggests that the irradiation under these conditions gives π-pulse.[16] The pulsed-LODESR spectrum at an average power of 10 W is shown in Figure 2a. Its FWHM was 0.14 mT (i.e., 3.9 MHz). A conventional LODESR measurement was also performed at a modulation frequency of 500 kHz, with the on/off ratio of 1:1 and an average power of 10 W. As a result, the LODESR spectrum where FWHM was 0.06 mT (i.e., 1.7 MHz) was obtained (Figure 2b). The FWHM of the pulsed-LODESR spectrum is 2.2 MHz wider than that for the conventional LODESR, meaning that pulsed radiowaves irradiate over the bandwidth of 2.2 MHz, approximately. This value is smaller than the theoretical value (4.3 MHz) due to some reasons (for example, pulse shape is not strict rectangle because of Q>1). However the bandwidth of 2.2 MHz is wider than the FWHM of the TAM solution (1.7 MHz).

In this study, a new type ESR resonator for pulse techniques was presented. Its Q is sufficiently low so that irradiation is possible over a wide frequency band. Furthermore, it can handle a sufficiently high power according to the *in vivo* sample size. It is believed that this resonator will aid in the development of *in vivo* pulsed RF ESR spectrometers.

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14 The authors thank Professor K. Golman, Nycomed Innovation, Malmo, Sweden, for the generous gift of the spin probe used in this study.
15 This value (670 ns) was obtained from 1 mmol dm^{-3} aqueous solution of TAM by employing an X-band pulsed ESR spectrometer (RE-3X equipped with ES-PX1000, JEOL). In an equation, $y=e^{(-x/a)}$, when a value of 200 ns or 670 ns was used for *x* or *a*, respectively, y=0.74 was obtained. Thus we believe that a pulse width of 200 ns is sufficient narrow for the TAM solution.
16 Alternating magnetic field (B_1) for π-pulse measuring 200 ns in width is estimated to be 89 μT from the pulse width and g value. When a maximum power of 100 W is irradiated to the resonator, the B_1 in the resonator is estimated to be 184 μT from the Q value, dimensions of the resonator, and applied power. These B_1 values are roughly similar.