Spatial distribution of free radicals in dental resins using Electron Paramagnetic Resonance Imaging (EPRI)

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Introduction & Aim of the study

Methacrylate resins are highly popular in dentistry and are largely used in clinic for tooth restoration. Free radicals are created during the photopolymerization process and can be detected by EPR spectroscopy. Two main radical species have been identified, an allylic radical and a so-called "propagating" radical, giving a complex multiple lines EPR spectrum (Fig.1). EPR spectroscopy was used to monitor the evolution of the global radical concentration, involved in the well-known post-polymerization shrinkage phenomenon[a].
EPRImaging allows to study the spatial distribution of free radicals and would constitute an attractive alternative method to study the regional evolution of radical distribution in biomaterials such as dental resins. Nevertheless EPRI requires spectral characteristics to be satisfied in order to build an artifact-free and non biased image. Shortly, the complex signal must be deconvoluted before image reconstruction, and the linewidth of the signal must not vary inside the object. This study focuses on these two points and explores the main characteristics of the acquired image. The evolution of the main radical species is also observed in a small phantom made of experimental dental resin using 2D spectral imaging.

Material & methods

Two types of resins were used. The commercial resin was Charisma® (A3, Heraeus Kulzer) composed of 36 % (vol) of organix matrix and 64 % (vol) of mineral filler. An experimental transparent resin containing no mineral filler was also used (mix of Bis-GMA (Heraeus Kulzer) and TEGDMA (Aldrich). Resins were molded using an empty 1ml PE syringe, the samples were photo polymerized with the light provided by a Translux Energy device (Heraeus Kulzer) positioned at the top of the sample. Two different energy settings were used (high & low), with an illumination time of 20 sec.
Imaging was performed on a Bruker Elexsys E540 system operating at 9 GHz, equipped with a Super High Sensitivity Probe. In a first experiment, 2D spectral imaging (2DSI) was used in order to check the evolution of the linewidth within the sample. No deconvolution was applied in 2DSI. Classical 2D images were then acquired, with deconvolution applied before image reconstruction, in order to test the robustness of the process. 2DSI was again used to study the spatial distribution of the two radical species.

Results

In 2DSI, all the peaks displayed a constant linewidth throughout the sample. Because there was no variation in LW, deconvolution could be applied to conventional 2D images. These images reconstructed after deconvolution reflect the density of radicals. It decreased from the top of the sample (were illumination occurred) to the bottom where the intensity was the lowest. Differences in radical distribution were also noted according to the type of resin used and the power of illumination (Fig. 2). Using 2DSI it was possible to measure the intensity of each peak in each pixel and build a map of the signal for each radical species. A significant decrease in signal intensity from top to the bottom of the sample was observed with the allylic radical.

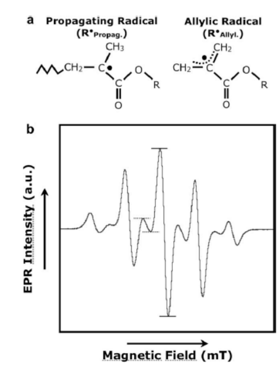

Fig. 1: complex EPR spectrum

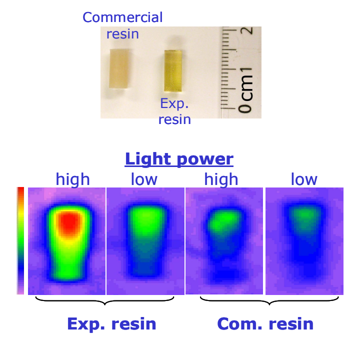

Fig.2: 2D EPRI images obtained after deconvolution of the complex spectrum.

Conclusion

Although dental resins exhibit a complex EPR signal, theoretically unfavourable for EPRI, it was nevertheless possible to acquire image of good quality because of the very strong signal and of a constant linewidth. The images reflect the spatial distribution of free radicals in dental methacrylate resins. This method, together with 2DSI, offers the unique possibility of non destructive characterization and mapping of free radicals in biomaterials and materials science.

References

a. Leprince J, Lamblin G, Truffier-Boutry D, Demoustier-Champagne S, Devaux J, Mestdagh M, Leloup G. Acta Biomater. 2009 Sep;5(7):2518-24