This paper describes three methods for making ultrahomogeneous ceramics and glasses by utilizing mixing in solution instead of the traditional mixing of powders. It is the first review of the sol-gel method that we developed, including the use of organic precursors, for making ceramics. [The SCI® indicates that this paper has been cited in over 135 publications.]

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I developed the solution-sol-gel method because I was lazy and was always looking for simpler ways to accomplish something. From 1946 to 1948, as a dutiful graduate student under E.F. Osborn, I was engaged in making really homogeneous glasses or ceramics for the precise study of fundamental phase equilibria in high-temperature oxide systems. The accepted tradition, developed over decades at the Carnegie Institution of Washington, was to mix fine oxide powders (note that ~325 mesh still means 450,000 Å particles) tediously under alcohol in a mortar; to transfer all the contents (hardly easy) to a platinum crucible; to melt the contents at 1400°-1700° C (depending on the composition) for an hour or two; and then to quench the contents in water. Whenever a glass formed, it had to be cracked out of the crucible with a special hammer over glazed black paper. Every tiny shard was scraped up. The glass was then crushed in a steel mortar. Iron particles were then removed from the crushed glass by careful and imaginative use of magnets, and the powder was restored to the crucible and remelted and quenched. The entire process had to be repeated three to five times to get really homogeneous ceramics. I probably prepared 100-150 compositions in this way just to obtain starting materials for my PhD work.

It is therefore little wonder that when I started as Osborn’s postdoc in July 1948, my first goal was to find a better method to make homogeneous ceramics. As an inorganic physical chemist I reasoned that putting all the ions into solution reduced the mean separation of the constituents by 10^4. The difficulty in making silicates, of course, was the fact that Si could not be put into solution without simultaneously adding other inorganic ions such as Na. I therefore turned to organic precursors. Tetraethoxysilane was easy to obtain, but in 1948 finding or making the other metal organics (for Al, Ti, and so on) was hardly simple. The method of mixing these organics in an alcohol solution, hydrolyzing to a sol and setting to a gel, and firing at 500°-700° C to a homogeneous ceramic powder proved to be a universally applicable process. It was so simple that it was hardly worth describing in great detail; hence the brevity of the paper.

In the following decade a dozen PhD students of mine probably made several thousand homogeneous compositions including all of the most basic systems of ceramic science and technology via the sol-gel route. The method became routine in geochemical research worldwide. In spite of repeated presentations at technical meetings, 10-15 years passed before the first technologies (for example, nuclear fuel pellets) based on the sol-gel method appeared. And another 10-15 years passed before the sol-gel method reappeared in ceramic science as a new fashion. By that time this paper and the three dozen others we wrote on sol-gel ceramics before 1960 were “occluded”; they are rarely cited by the majority of US ceramics and glass researchers since no other university ceramics group did any work in the field until about 1975. In 1982 I reentered sol-gel research, reversing my original goal by instead making ultra-homogeneous ceramics via multiphase xerogels.