

Efficiency enhancement of decontamination gels by a superabsorbent polymer



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HIGHLIGHTS

- We studied how the introduction of a superabsorbent polymer in alumina decontamination gels enhances their efficiency.
- Ions dissolved near the contaminated surfaces make the polymer beads to unswell via an osmotic pressure effect.
- The beads size variations modify, sometimes badly, the overall rheology.
- An optimal formulation can be found, averaging the two effects.

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ABSTRACT

Physical gels have been studied as an advantageous alternative to traditional decontamination techniques, which most often rely on the use of surfactant solutions. Such physical gels consist of colloidal particles suspended in a decontaminating solution. They can be sprayed over the surfaces to be treated, where they dry and form solid disposable residues. In order to improve their efficiency, the formulation of gels containing superabsorbents (such as sodium polyacrylate) has recently been suggested. But the mechanism accounting for the observed efficiency increase has not yet been investigated. These additives have a large swelling potential in aqueous media and the amount of adsorbed water depends on the presence of salts in the medium. We anticipated that specific ion effects on the swelling behavior of superabsorbent polymers were the driving force of efficiency enhancement and confirmed this assumption by rheology and in situ tests.

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1. Introduction

Decontamination is a major task of nuclear plant decommissioning operations. Decontamination also must be undertaken very rapidly in the eventuality of CBRN (chemical, biological, radiological, and nuclear) terrorist attacks in public buildings. Although the context and intervention conditions in these situations are completely different, the aim of decontamination is the same in both cases: protect the health of the population and enable further utilization of the concerned buildings. Not only the aim is the same, in most cases the decontamination techniques are very similar and still are a subject of innovation. In addition to classical techniques such as high pressure surfactant solution or superheated steam

cleaning, more advanced techniques have been proposed recently, including the use of foams, CO₂ ice blasting, or laser [1–3]. Among these advanced decontamination techniques our laboratory has been working on physical gel-based approaches for nuclear, biological, and chemical decontamination [4–6]. Such gels are formulated by suspending mineral colloids (e.g. silica or alumina) in an aggressive solution, together with suitable additives (e.g. surfactants). Depending on the application, the aggressive solution may be acidic and oxidizing (e.g. nitric acid containing solubilized cerium IV) in order to corrode metallic surfaces and remove radionuclides, or strongly basic (up to 5 M NaOH) when bacteriological decontamination is required. In industrial applications the formulated gels are sprayed over contaminated surfaces. The drying time must be slow enough to enable the active species of the gels to chemically attack the surfaces and/or contaminants. After drying, they form non-powder solid residues, which are easy to handle and dispose. Fig. 1 shows a schematic view of the process.

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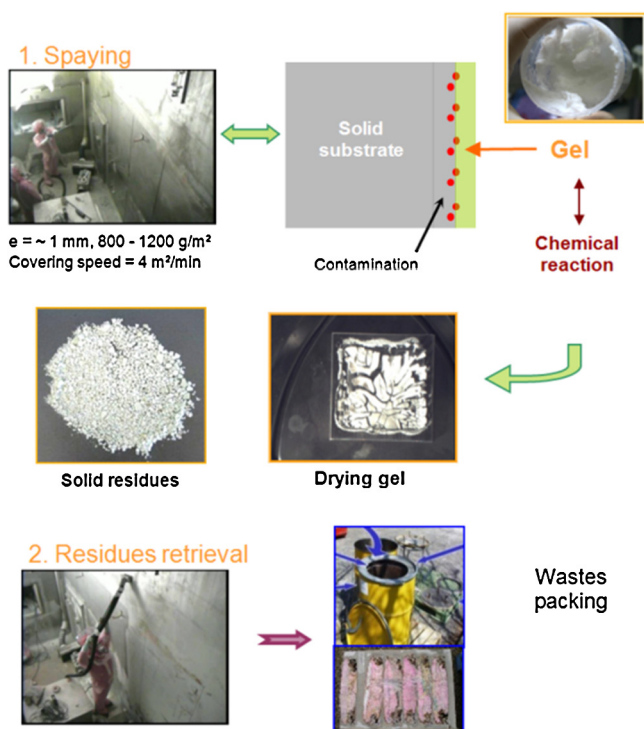


Fig. 1. Decontaminating gel process.

As this description suggests, the rheological behavior of these gels is a key feature in the process. It plays a role in the spraying step and implies how they will behave on the substrates. Indeed, characteristics such as low viscosity for spraying and sufficient yield stress for the gel to remain on surfaces are mandatory and must be measured. Obviously, good thixotropic behavior is also required for the gels to recover their characteristics after spraying. Typical values are a viscosity ranging from about 10^6 Pa s at low shear rates to 10^{-2} Pa s at high shear rates ($\sim 10^6 \text{ s}^{-1}$) and a yield stress of about 50 Pa (values from our own measurements). If these requirements are not achieved, a bad surfaces covering could follow, leading to an insufficient decontamination. Moreover, for optimization concerns, the “strength” needed for pulverization is important. It is then of utmost importance to conduct a rheological study, at least regarding the process parameters. Fortunately, the physical gels used here exhibit characteristics which fit process requirements thanks to their own structure: particles linked by weak electrostatic forces that can organize when they flow, lowering the overall viscosity of the material as the shear rate increases. Chemically speaking, we seek reagents that can attack the treated surfaces to a depth of several micrometers. The radioelements are then naturally dissolved. However, the efficiency greatly depends on the surface material, and the reagents used can influence the gel structure, as we will discuss later.

Some additives have been recently introduced in order to change the existing properties of the gels or to develop new process variations. Among them, it has recently been suggested that the use of superabsorbent polymers (SAP, e.g. sodium polyacrylates) enhance the decontamination efficiency, especially on partly porous substrates such as concrete [4]. The origin of this effect, however, remained unclear and it is the goal of this contribution to shed some light on the chemical effects accounting for these observations and to highlight how they modify alumina suspensions behavior.

SAP are widely employed in industry for their ability to absorb large amounts of liquid [7,8]. Most of them are based on polyacrylate polymers (polyacrylic acid–sodium polyacrylate)

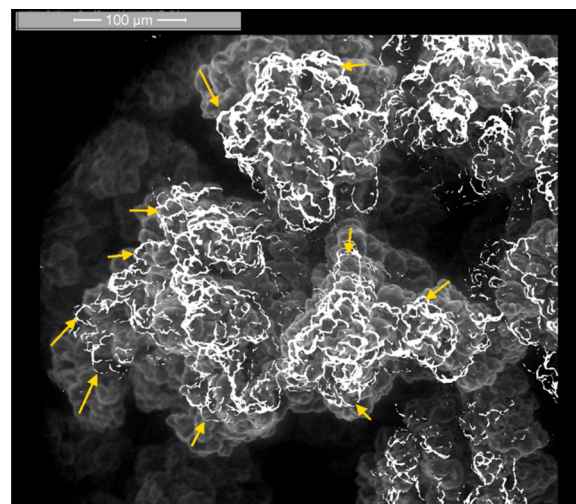


Fig. 2. ESEM images of NaPAA beads; HR = 95% (background image) and 50% (bright shape).

even though xanthan–PVA (polyvinyl alcohol) and other formulations can be found [9,10]. In the case of sodium polyacrylate ($[\text{CH}_2-\text{CH}-(\text{COONa})-]_n$) or “NaPAA”, it is known that the polymer chains are coiled in the dry state. When exposed to moisture, the solubilization of Na^+ ions increases the repulsion between the negatively charged carboxyls. As the polymer is weakly cross-linked, osmotic pressure enables water to further enter the polymer network until a gel-like material is formed [11]. This is evidenced in Fig. 2, showing our own images obtained on an environmental scanning electron microscope (ESEM). The figure shows the different volumes of a gel at 50% and 95% relative humidity. Bright arrows highlight the dimensional changes.

It has, however, been observed that the presence of salts in the aqueous media strongly impacted the degree of expansion of the polymers. As we work on cement (or also metallic) substrates, a certain amount of dissolved ions may come into contact with NaPAA; the potential effects must then be taken into account. This unswelling phenomenon has been investigated in various studies, either considering “isolated” chains whose hydrodynamic radii change [12–14] or the whole structured gel [15,16]. In such systems, the swelling equilibration is given as a combination of elastic behavior, chain/solvent mixing behavior and an osmotic effect due to the presence of counterions in solution. Some studies insisted that the main parameters controlling the swelling of the gel were osmotic pressure and the mixing contribution [17,18] for monovalent or divalent cations [19]. Molecular dynamic simulations precise this outcome [20]. The solution pH plays a part in the swelling behavior: a reversible contraction is observed between pH 3 and pH 13 with an optimum value at pH 6 where the bead volume changes the most. Below pH 3 the process is irreversible as protons replace the sodium in the gel [21]. Finally, isolated polyacrylate chains can also be adsorbed on particle surfaces in mineral slurries, modifying their overall viscosity [22–24].

In the following, we will discuss how the swelling behavior of alumina/NaPAA gels affects decontamination processes. More than a simple efficiency enhancement, it has also heavy consequences on how the gels behave rheologically and it is thus important to precisely describe the mechanisms that can affect how they are handled in the decontamination process (viscosity changes, behavior with the contaminated surfaces...). In top of that, the materials we studied are, apart from their industrial use, colloidal gels in which some particles can endure morphological changes. Consequently, particles’ jamming is observed and a secondary network made by the polymer beads appears within the colloidal

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