



# Formulation of new materials based on geopolymer binders and different road aggregates

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## Abstract

The study of bitumen substitution in the field of road construction is necessary to overcome the effects of limited oil resources and economic problems. The elaboration of new materials based on geopolymer binders and aggregates was investigated in this work. Two series of formulations of geopolymer binders based on sodium or potassium were prepared with the incorporation of granite or diorite. The formulation of these binder was investigated by the study of (i) the effect of the nature of the alkaline solution by modify the pH value with base and (ii) the mass percentage of the incorporated aggregates. The time evolution study shows different aspects of the synthesized materials, which depend essentially on the composition of the activating solutions and the nature of the alkaline solutions. The formulations using these ratios ( $\text{Si/Na} = 1.55$ ;  $\text{Si/K} = 1.94$  and  $2.96$ ) appear the best. Moreover, it was evidenced that materials based on the potassium alkaline solution present a consolidated aspect faster than the materials based on the sodium alkaline solution. Additionally, it is possible to incorporate diorite or granite until 60 w% with some formulations. These various formulations are promising to be use not as a substitute of bitumen but in addition between bitumen and ground.

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## 1. Introduction

In the road coating field, bitumen, produced by refining crude oil, is the oldest and leading material used in road construction and various types of pavements [1]. The current world consumption of bitumen is approximately 102 million tons per year according to the Asphalt Institute and Euro-bitume in 2011 [2]. However, with the growth of environmental awareness and the limited oil resources for producing high-quality bitumen, as well as economic consideration, research is oriented toward the modification or the substitution of bitumen [3–5]. Several materials such as biomaterials, eco-materials and raw materials (wood or palm lignin, resin, etc.)

can be used as substitutes for bitumen because they present the same characteristics (viscoelastic and rheological properties, adhesion, hydrophobicity [6–8]) and even more advantages such as small-scale production, the decrease of road maintenance and required transportation. During the last 40 years, an increasing number of researchers have focused on the modification of bitumen to enhance its quality [9] and also on the substitution of the bitumen by the use of geopolymer [10].

Polymer modification of bitumen by mechanical mixing and chemical reaction [11] were used to improve some properties of the bitumen such as yielding a higher stiffness at high temperatures, higher cracking resistance at low temperatures, better moisture resistance or longer fatigue life with preserved adhesion and viscoelastic properties [12–14]. The main disadvantage of these types of modification is the incompatibility between the polymer and the bitumen in terms of the chemical

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structure and the reactivity of polymers, which depend on their respective organic and inorganic composition [15]. However, “geopolymers” which present similar rigidity and resistance to alkalinity as bitumen, are considered to be promising materials to substitute bitumen. These materials belong to a family of aluminosilicate, synthesized by the reaction between an oxide aluminosilicate and an alkaline solution at room temperature. They present promising qualities such as a lower calcination temperature during their production; less CO<sub>2</sub> emission; reasonable strength, which can be gained at room temperature in a short time; resistance to fire and acid attacks; low permeability; capacity to attain higher unconfined compressive strength; and much less shrinkage than that of Portland cement [16–20]. Most of the studies [21,22] focus on evaluating the properties of formulations of geopolymers by determining the impact of the nature of the silica source, the alkali concentration, the Si/Al ratio, and the alkali/fly ash ratio. They show that geopolymeric cement made with a reasonable mix-design and formulation can exhibit properties that are superior to those of Portland cement, making it a strong substitution candidate for Portland cement in the fields of civil, bridge, pavement, hydraulic, underground and military engineering. According to the literature, the metakaolin is considered the main precursor in the formation of geopolymeric materials due to its high reactivity and purity compared to the other materials [26]. The properties of these geopolymeric materials depend on the aluminosilicate source and the alkaline solution used [23–27]. In fact, the Si/Al, Si/M and Si/H<sub>2</sub>O ratios play a primordial role in the polymerization degree and the structure of the synthesized materials [27–29].

Despite of the chemical, mechanical and microstructural studies of geopolymers, slight works have been done on the materials based on geopolymers and aggregates. Wang et al. [30] studied the microstructure and the bonding strength of the interface between marble and granite aggregates on slag-based geopolymer. The results show that the bonding strength of geopolymer–marble interfacial transitional zone was higher than that of geopolymer–granite. Indeed, with the development of hydration, the compact block structures were formed and connected to aggregates on the surface of geo–marble/geo–granite interfacial transitional zone. However, Borges et al. [31] investigated in their work the use of materials based on geopolymers with quartz and glass as aggregates for floor tile. The results show that the materials based on quartz present higher compressive and flexural strength and lower porosity. Indeed, Kamseu et al. [32] showed in their studies that the use of fine aggregates such as ladle slag, nepheline syenite and quartz sand can improve the interfacial zone between geopolymer and aggregates which is important for the design and the production of dense, resistant and durable composites. Also, the studies of Arellano-Aguilar et al. [33] highlight the importance of the determination of the optimum balance between aggregate and binder content as well as the composition of the binder to produce geopolymers with suitable properties.

The aim of this work is to synthesize new materials based on geopolymer binders and road aggregates (diorite and granite) as strengtheners. The effects of the cation nature (Na or K) and the quantity of NaOH and KOH on the feasibility of the

Table 1  
Characteristics of the raw materials.

Nature	Supplier	Weight chemical composition (%)				
		SiO <sub>2</sub>	M <sub>2</sub> O (MO: Na or K)	H <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	Impurity
SiO <sub>2</sub> 3.4Na <sub>2</sub> O	Woellner–France	27.5	8.3	64.2	–	–
SiO <sub>2</sub> 3.4K <sub>2</sub> O	Chemical labs	16.4	7.6	76.0	–	–
Metakaolin	Imerys–France	55.0	–	–	40.0	5.0
Diorite (6/10 mm)	Colas–France	–	–	–	–	–
Granite (6/10 mm)		–	–	–	–	–

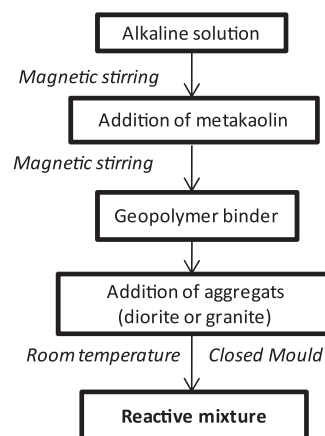


Fig. 1. Synthesis protocol of the different formulation.

geopolymer binder were investigated. Indeed, the impact of the mass percentage of the incorporated aggregates was studied to determine the ideal formulation of the consolidated materials.

## 2. Materials and methods

### 2.1. Materials

In this study, two alkaline silicate solutions were used (sodium and potassium), as well as the metakaolin M-1 as an aluminosilicate source. Two types of road aggregates were incorporated (grain size 6/10 mm) with different percentages. Table 1 presents the characteristics of all the raw materials used.

### 2.2. Samples preparation

Fig. 1 presents the synthesis protocol. The mixtures were prepared in the first step by dissolving KOH pellets (85.7% of purity) or NaOH pellets (99.0% of purity) in potassium silicate (Si/K=1.7, 76.0% water) or sodium silicate (Si/Na=1.7, 64.2% of water) solutions. Then, metakaolin was added to the alkaline solution under magnetic stirring to form the geopolymer binder. In the second step, two types of road aggregates were incorporated

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