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BIOMIMETIC MATERIALS: GEOPOLYMER-BASED CONCRETE, INSPIRED BY THE PROPERTIES OF SPIDER SILK.

***Abstract*— This research project presents a review of biomimetic materials, specifically bio-inspiration in the properties of spider silk, applied to the development of higher quality concrete. The use of ordinary Portland cement in concrete poses an environmental problem due to the emissions generated during its processing. Using a mixed approach, with numerical data to quantify concrete strength and qualitative data on concrete properties and concepts, data was collected from different articles, evaluating various aspects of the mixtures, resistance to different stresses, workability, and chemical composition, to visualize how this affects the final product. The mixtures with the best results were separated, these being those that incorporated blast furnace slag, but it was noted that several of these mixtures presented other problems, so those that incorporated fly ash were also analyzed, this being a good combination for this type of concrete, as a good incorporation of both gives better results in workability, strength, and durability, but this also depends on the content of silica, alumina, and calcium that it contributes to the total mixture. The mixtures that gave the best results in terms of strength were observed in a mixture of blast furnace slag, silica fume, and coal ash, with strengths of 90.5 in compression, 7.8 in tension. In terms of workability, the best results were obtained with a mixture of fly ash and the other normal concrete aggregates, with a slump of 180 mm.**

Keywords*— *geopolymer concrete, fly ash, blast furnace slag, biomimicry, dragline spider silk

I. Introduction

Nature has always demonstrated its great ingenuity by creating structures that solve problems in amazing ways. Currently, one of the biggest problems we face is pollution caused by industry, especially the construction industry, which uses materials that are harmful to the planet, as well as to the people and animals that inhabit it. In 2022 alone, 2.41 gigatons of carbon dioxide were emitted from cement production [1]. Therefore, one way to solve these problems is to see how nature itself solves them.

This is where biomimicry comes in, which means "imitation of life." For the purposes of this research, spider silk is being used as the natural material, and concrete will be used as the artificial material. The source of inspiration here is that spider silk is a very resistant material. Considering its density, it is capable of withstanding great stress, which is a characteristic sought after in concrete. In addition, by drawing inspiration from natural fibers, which are less harmful than the components of cement.

The research will analyze mixtures of different materials to find patterns that provide the best resistance and compare them with control mixtures for traditional concrete, thus defining better methods of analysis before physical testing.

All this with the aim of finding alternatives that help to reduce, partially or totally, the high consumption of ordinary Portland cement, obtaining concrete with better physical characteristics and more environmentally friendly.

II. METHODOLOGY

This study follows a mixed-methods approach, using qualitative data for a better understanding of the concepts, applications, and characteristics of biomimicry and geopolymers; and quantitative data to explain characteristics such as tensile strength, compressive strength, flexural strength, among others, and the chemical composition of concrete mixtures.

A. Variables

A dependent variable was taken from four independent variables. This variable is: The properties of bio-inspired concrete, based on geopolymers. The independent variables are:

- Properties of spider silk: its chemical structure and mechanical properties.
- Design of polymer mixtures: chemical composition, base material, and alkaline activator used, as well as extra aggregates.
- Mechanical properties of the tests: their mechanical resistance to different types of stress.
- Reduction in cement use: The amount of cement use that can be reduced with geopolymers.

19 *B. Methods*

- Use of Thesauri: This technique allows users to find synonyms or search terms relevant to the topic, which helps to expand the search results related to the topic of interest.
- Filtered and updated search: Specific keywords will be used to perform searches, in addition to filtering the results so that they are open access and scientific articles, as well as recently published, no more than 5 years old, which will facilitate the search for more relevant and up-to-date material.
- Compilation tables and matrices: Use tables and matrices to compile information and analyze and visualize it in a better way.

C. Methodological process

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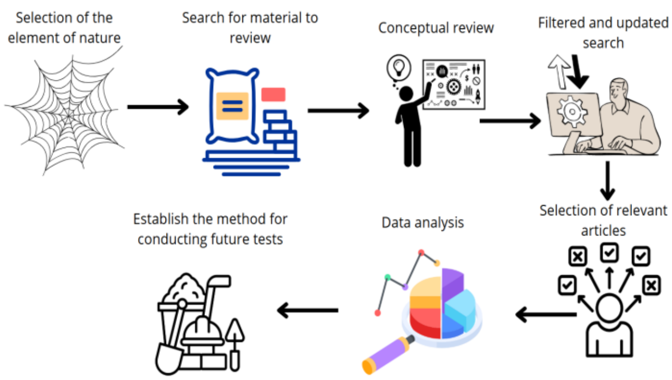

Fig. 1. Procedure performed to obtain results.

Figure 1 shows the methodological process to be followed to obtain the results. To obtain the results, the element of nature that serves as inspiration is initially chosen, in this case spider silk, specifically "major ampullary silk." The material that could mimic its properties is sought, namely geopolymers. To better understand what is being researched, a specific conceptual investigation of biomimicry, geopolymers, and mechanical resistance is carried out. Subsequently, a filtered search is performed, using thesauri, and updated, with articles no older than 5 years. From the articles found, the most relevant ones are selected, data is extracted from them, and tabulated, such as chemical composition, mixtures, resistances found, and workability, in order to better analyze the reason for their properties, how they can be compared to spider silk, and reduce the use of cement. Finally, a method for conducting subsequent physical tests is sought.

D. Mixture Identification Codes

Table I explains the meaning of the codes used to identify the mixtures studied, listing the mixture number and the article in which it can be found.

2

40 TABLE I. MIX NOMENCLATURE

Code	Description	
	Number of mixture	*Article*
AR-6-2	2	[5]
AR-7-5	5	[6]
AR-3-1	1	[7]
AR-10-1	1	[13]
AR-8-1	1	[14]
AR-11-4	4	[15]
AR-6-4	4	[5]
AR-4-3	3	[17]

III. Analysis and Results

A. Properties of spider silk

The spider can produce up to seven types of silk, of which the most interesting due to its excellent properties is the major ampullate silk, or dragline silk. This fiber is highly resistant to traction, even more so than Kevlar.

Spider silk is primarily composed of two spidroins, MaSp1 and MaSp2. These proteins feature both crystalline regions—formed by stacked β-sheet structures that provide tensile strength—and non-crystalline, amorphous regions, which include 3_{10} helices and β-spirals that contribute to elasticity. MaSp1 contains polypeptide motifs such as poly-alanine (An), glycine-alanine repeats (Gan), and GGXn (two glycines and a variable amino acid). MaSp2 includes these same motifs, in addition to GPGXXn (glycine-proline-glycine followed by two variable amino acids). The An and Gan sequences generate β-sheets associated with strength; GPGXXn forms β-spirals that enhance elasticity; and GGXn acts as a connector between rigid and flexible domains through 3_{10} helices [2] [3]

In 2021, a literature review [4] was conducted to characterize several mechanical properties of spider silk. The findings indicated a tensile strength of approximately 4 GPa, an elasticity of 35%, and a Young's modulus of around 10 GPa.

The most interesting thing about spider silk is its high tensile strength and toughness, which contrasts with concrete, which has low tensile strength and therefore needs to be reinforced with steel.

B. Chemical composition in the reviewed literature

For this review, studies conducted with aluminosilicate bases that are easier to find in the region will be used, prioritizing fly ash, followed by granulated blast furnace slag, as well as other aggregates to provide better tensile strength properties.

In this context, twelve experimental studies were identified in which the mixture compositions were provided, primarily using fly ash and ground granulated blast furnace slag as the main aluminosilicate sources. From a total of 55 different mixtures, the three with the highest compressive strength values were selected, since this is the most reported performance parameter in studies on this type of concrete. Additionally, the selected articles included the chemical composition of the materials used.

From these three main articles, the contributions of the key chemical components forming the aluminosilicates were extracted, and the results were compiled in tabular form.

For the first article [5], whose total density is 2476.5 kg/m^3, the weighted percentages are:

TABLE II. CHEMICAL COMPOSITION OF AR-6-2

Material	Chemical composition		
	Weighted SiO_2	*Weighted Al_2O_3*	*Weighted CaO*
Silica fume	4.53%	0.0046%	0.0139%
Blast furnace slag	3.28%	1.34%	4.31%
Total	7.8179%	1.3446%	4.3239%

For the second article [6] , whose total density is
10 2341.96 kg/m^3, the weighted percentages are:

TABLE III. CHEMICAL COMPOSITION OF AR-7-5

Material	Chemical composition		
	Weighted SiO_2	*Weighted Al_2O_3*	*Weighted CaO*
Fly ash	5.16%	2.16%	0.39%
Blast furnace slag	1.58%	0.60%	1.59%
Processed steel slag	10.77%	5.18%	10.26%
Total	17.53%	7.94%	12.24%

For the third article [7], whose total density is 2387.5 kg/m^3, the weighted percentages are:

TABLE IV. CHEMICAL COMPOSITION OF AR-3-1

Material	Chemical composition		
	Weighted SiO_2	*Weighted Al_2O_3*	*Weighted CaO*
Fly ash	11.28%	4.1%	1.61%
Total	11.28%	4.1%	1.61%

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Tables II, III, and IV show the individual chemical composition of each of the mixtures. Table V shows a summary of the above data, with the Si/Al ratio.

TABLE V. SUMMARY TABLE OF CHEMICAL COMPOSITION

Component	Summary table		
	AR-6-2	*AR-7-5*	*AR-3-1*
SiO_2	7.82	17.53	11.28
Al_2O_3	1.34	7.94	4.1
CaO	4.23	12.24	1.65
Si/Al	5.81	2.2	2.75

1

The mixture that gives the best results, AR-6-2, has a
35 high silica content compared to aluminum, with a Si/Al ratio of 5.81. The next mixture, AR-7-5 has a ratio of 2.20, and the AR-3-1 mixture has a ratio of 2.75. Therefore, it could be said that there is a certain tendency for greater resistance to be achieved as this ratio increases. This is because, with a higher silicon content, the C-S-H gel forms better, which gives greater strength. However, such low alumina content has consequences, as it prevents the other gels from forming properly, reducing the mixture's toughness. As for calcium, this component is lower in fly ash than in slag, which is interpreted as meaning that slag provides greater resistance, partly due to its high calcium content, but this affects its durability. This occurs because excess calcium forms a C-A-S-H gel (calcium, alumina, hydrates silica), , which does not have good resistance to high temperatures. When this happens, it loses strength and degrades. In comparison, the N-A-S-H (sodium, alumina, hydrated silica) gel can retain 63% of its original strength even at temperatures of 1173.15 K [8].

As for the Si/Al ratio, it is recommended that it be greater than 1.5, since when there is a greater amount of silica, the structure of the N-A-S-H gel is more stable, but this must also be controlled, since an excess of it, as can be seen in AR-6-2, can affect the long-term durability of the mixture.

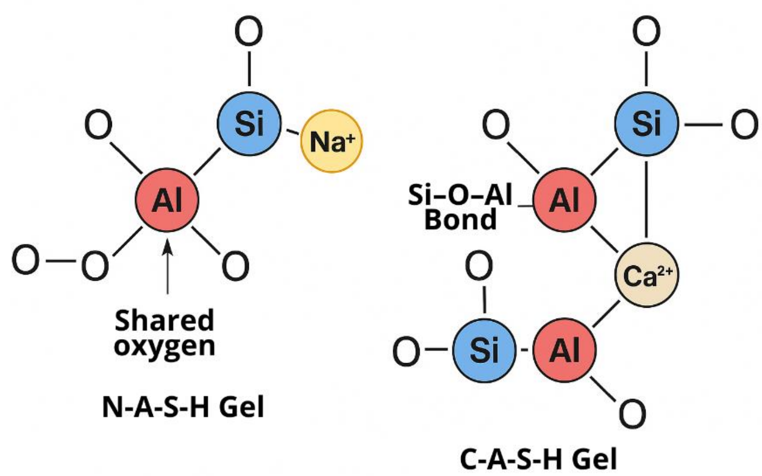

Fig. 2. N-A-S-H & C-A-S-H gel

Figure 2 shows the chemical structure of the gels formed in geopolymerization.

C. Workability

The workability of concrete is a very important factor when mixing, as concrete with very low workability is too dry, which affects its uniform pouring. Conversely, if it is too high, it affects its strength, as it is too liquid. According to the literature consulted, certain aluminosilicates provide too low workability, as is the case with sugarcane bagasse [7] or mixtures of blast furnace slag with fly ash and precipitated silica [9]. On the other hand, mixtures with coal ash and fly ash do provide good workability [5] The liquid/solid ratio L/S, to obtain better workability results, can be set between 0.22 and 0.40 [10].

Another study explains that the good workability of geopolymer concrete lies in the appropriate combination of alkaline activators, sodium silicate, and sodium hydroxide with water, together with fly ash, but when glass fibers are added, this workability is low [11].

D. Compressive strenght

Compressive strength is the most studied property in concrete, as it is very important for greater structural safety and for withstanding the loads to which it is subjected in different applications.

According to a review study conducted in 2021, chemical composition is one of the main factors responsible for the compressive strength of geopolymers, especially aluminosilicates, as they are necessary for the formation of NASH gel, which hardens concrete. This gel can be formed more effectively with higher temperatures and longer curing times [12].

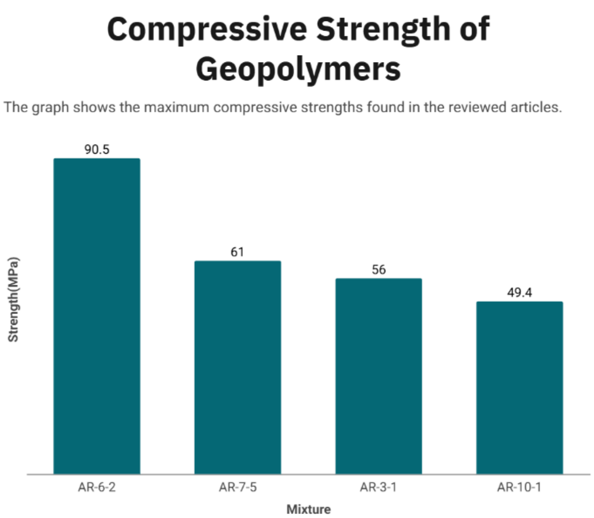

Fig. 3. comparison of compression mixtures

Figure 3 shows a comparison of the compressive strengths of different mixtures, measured in MPa.

The first mixture AR-6-2 contains 250 kg/m3 of blast furnace slag, 100 kg/m3 of silica fume, and 150 kg/m3 of coal ash. It is alkali-activated with 150 kg/m3 of sodium silicate and 75 kg/m3 of sodium hydroxide. The coarse and fine aggregates correspond to 1159 kg/m3 and 580 kg/m3, respectively. With a slump of 160 mm.

The second mixture AR-7-5 contains 100 kg/m3 of blast furnace slag, 233 kg/m3 of fly ash, and 584 kg/m3 of processed steel slag. Alkaline activated with 83.33 kg/m3 of sodium silicate and 33.33 kg/m3 of sodium hydroxide. The coarse aggregates correspond to 1155.1 kg/m3. No data is provided on its workability.

The third mixture AR-3-1 contains 500 kg/m3 of fly ash. It is alkali-activated with 105 kg/m3 of sodium silicate and 70 kg/m3 of sodium hydroxide. The coarse and fine aggregates correspond to 1000 kg/m3 and 700 kg/m3, respectively. Its slump is 183 mm.

The fourth mixture AR-10-1 corresponds to the control mixture, traditional concrete, containing 360 kg/m3 of Portland cement, coarse and fine aggregates corresponding to 1102 kg/m3 and 735 kg/m3 respectively, and 324 kg/m3 of water. Its workability is measured by a slump flow of 450 mm.

All geopolymer mixtures give better compression results than conventional concrete, and that mixtures with blast furnace slag have greater strength than those with fly ash. However, their workability is lower, so a mixture of both is better.

E. Tensile strength

The tensile strength of concrete is quite low compared to its compressive strength, which is one of its weaknesses. That is why it is reinforced with steel, such as rods. However, with advances in geopolymers, adding fibers can improve these results, although they still fall short of those exhibited by spider silk.

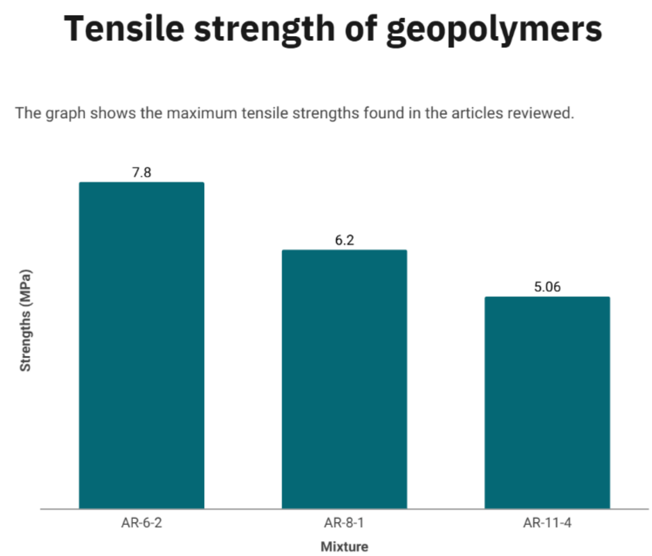

Fig. 4. comparison of tensile mixtures

Figure 4 shows a comparison of the tensile strengths of different mixtures, measured in MPa.

The first mixture AR-6-2 contains 250 kg/m3 of blast furnace slag, 100 kg/m3 of silica fume, and 150 kg/m3 of coal ash. It is alkali-activated with 150 kg/m3 of sodium silicate and 75 kg/m3 of sodium hydroxide. The coarse and fine aggregates correspond to 1159 kg/m3 and 580 kg/m3, respectively.

The second mixture AR-8-1 contains 375 kg/m3 of fly ash. It is alkali-activated with 168 kg/m3 of sodium silicate and 32.1 kg/m3 of sodium hydroxide. The coarse and fine aggregates correspond to 1124 kg/m3 and 607 kg/m3, respectively.

The third mixture AR-11-4 contains 128 kg/m3 of blast furnace slag and 297 kg/m3 of fly ash. It is alkali-activated with 143 kg/m3 of sodium silicate and 57 kg/m3 of sodium hydroxide. The coarse and fine aggregates correspond to 1124 kg/m3 and 607 kg/m3, respectively.

In terms of tensile strength, it can be observed that geopolymers improve this strength to a certain extent, given that normal strength, without reinforcement, is considerably lower than that observed in the studies investigated.

To improve tensile strength, certain natural fibers, properly treated, can be added, such as sisal fibers, which can increase tensile strength and modulus of elasticity by up to 70% [16].

F. *Flexural strength*

Flexural strength is the ability to resist forces that cause concrete to bend. To compare this in the review, the three best results for strength in 28 days will be selected.

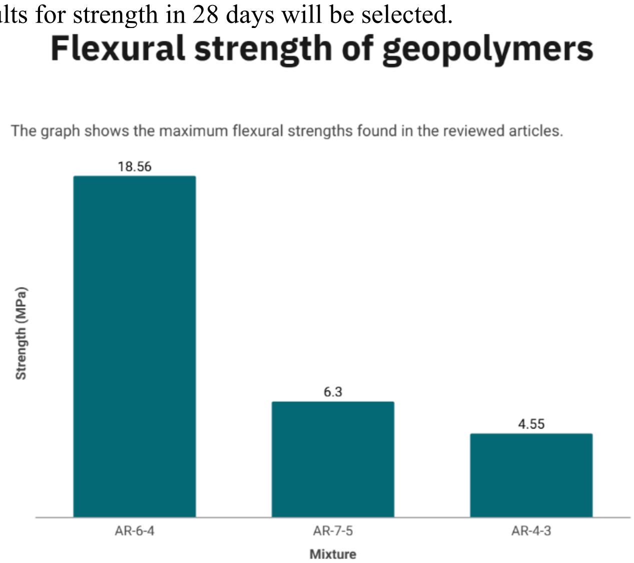

Fig. 5. comparison of flexural mixtures

Figure 5 shows a comparison of the flexural strengths of different mixtures, measured in MPa.

The first mixture AR-6-4 contains 250 kg/m3 of blast furnace slag, 100 kg/m3 of coal ash, and 150 kg/m3 of fly ash. It is alkaline activated with 150 kg/m3 of sodium silicate and 75 kg/m3 of sodium hydroxide. The coarse and fine aggregates correspond to 1160 kg/m3 and 580 kg/m3, respectively.

The second mixture AR-7-5 contains 100 kg/m3 of blast furnace slag, 233 kg/m3 of fly ash, and 584 kg/m3 of processed steel slag. Alkaline activated with 83.33 kg/m3 of sodium silicate and 33.33 kg/m3 of sodium hydroxide. The coarse aggregates correspond to 1155.1 kg/m3.

The third mixture AR-4-3 corresponds to the control mixture, traditional concrete, containing 308.39 kg/m3 of Portland cement, coarse and fine aggregates corresponding to 1178.5 kg/m3 and 767.74 kg/m3 respectively, and 138.64 kg/m3 of water.

G. *Optimal mixture designs.*

When it comes to a single dosage, several aspects must be considered, as a major limitation is that each aluminosilicate may contain chemical differences. That is why it is important to first analyze the binders and verify that they meet the requirements of ASTM C989 for blast furnace slag and ASTM C618 for fly ash.

In Honduras, the limited availability of materials is a major constraint on the use of this type of concrete. The easiest material to obtain is fly ash, a by-product of biomass combustion plants. Alkaline activators can be obtained from specialized laboratories, and aggregates, both coarse and fine, from hardware stores. In the case of blast furnace slag, this is an imported product, so economic viability is something to be reviewed.

A balanced mixture should contain fly ash and blast furnace slag in similar proportions. For laboratory tests, use dosages ranging from 35/65 for both aluminosilicates, since a high percentage of blast furnace slag offers greater strength due to its higher calcium content, but its workability is not the most efficient. On the other hand, fly ash, which has a higher percentage of silica and alumina, contributes to greater workability, although it provides less resistance. When activated with sodium hydroxide and sodium silicate as alkaline activators, it contributes more effectively to the creation of N-A-S-H gels, the main geopolymer gel. In addition, to gain tensile and flexural strength, adding fibers such as sisal or glass, in the range of 1% to 3%, helps in this regard.

In mixtures, it is important to pay attention to the L/S ratio, as the final characteristics of the concrete also depend on it. As mentioned in section 5.4, this value can be between 0.22 and 0.4 to obtain mixtures with an acceptable liquid content (this liquid can be water and alkaline activators). When the mixture contains a lot of water, it is more workable and more efficient to pour, but it greatly affects the strength. Conversely, when the mixture is too solid, it provides greater strength, but its workability is low, and pouring errors can occur [12].

Chemical analysis of the components is vital when making mixtures, as the Si/Al ratio is important. It is recommended that this ratio be greater than 1.5 but less than 3.0, as a high silica content improves strength but reduces toughness. Conversely, if the percentage of alumina is higher, the strength will not be as good.

IV. Conclusions

Although geopolymer mixtures exhibit very good compressive strength and increase tensile strength, they cannot compare to the strength of spider dragline silk. However, the use of geopolymers improves this property, and if lighter aluminosilicate materials are investigated, it is possible to achieve an imitation of a lighter and stronger concrete, like spider silk.

2 The geopolymer mixtures that give the best results are those containing blast furnace slag mixed with other aluminosilicates, such as coal ash, achieving strengths of up to 90 MPa. However, these concretes have low toughness and may be affected in the future due to their high calcium content. The addition of fly ash stabilizes them better.

Correct dosing is important to achieve better properties, as the quality of concrete depends not only on its strength but also on its workability, so it is vital to find the perfect ratio for it. Some aluminosilicates, such as fiberglass, provide excellent strength but very poor workability when too much is added, which compromises the correct pouring of the concrete.

In most of the mixtures analyzed, which give the highest strength results in terms of compression, traction, and flexural strength, Portland cement has been replaced 100%. This raises the possibility of a future in construction where CO2 emissions from clinker production are much lower than they are today.

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