

Application Properties of Bath Liquids for Children Based on Sodium Laureth Sulfate with Addition of Different Molecular Weight Collagens Derived from Marine Sources

Emilia Klimaszewska¹  · Marta Ogorzałek¹  · Artur Seweryn¹  · Tomasz Wasilewski¹

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Abstract This article investigates the influence of marine collagen of various molecular weights (Collagen Amino Acids—150 Da, Hydrolyzed Collagen—12,000 Da and Soluble Collagen—300,000 Da) on the functional properties of bath liquids for children based on anionic surfactants—sodium laureth sulfate. In addition to the aspects related to safety-in-use, which were presented in the authors' first work about marine collagen, consumers also expect bath cosmetics for children to have specific functional characteristics including viscosity or foaming properties. An increase in the molecular mass of marine collagen was found to be accompanied by an increase in dynamic viscosity and foaming ability and a decrease in the ability to emulsify fatty soil in the formulations under study. It can, therefore, be concluded that the addition of the highest molecular weight of marine collagen (300,000 Da) to bath liquids for children contributes not only to reducing the irritant effect caused by anionic surfactants, but also improves the usable properties of these types of cosmetics.

Keywords Anionic surfactants · Marine collagen · Bath liquids for children · Functionality properties

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Introduction

Bath liquids for children in physicochemical terms are aqueous solutions of surface-active compounds (anionic,

nonionic, and amphoteric) and various types of additives (rheology modifiers, fragrance compositions, humectants, emollients, preservatives, dyes, pearlescent agents, pH regulators, *etc.*). Due to the fact that surfactants, in particular anionic, can cause skin irritation, development trends in these types of products focus on the use of raw materials that will be safe and gentle on the child's skin. It should be noted that it differs significantly from the skin of an adult. The ratio of skin surface to body weight is 2–3 times greater than that of an adult. Thus, the amount of potentially absorbed substances in the child's skin is higher in correspondence with the adult body weight (Barel et al., 2014; Gelmetti, 2001; Walters et al., 2008).

To reduce the irritation potential of cosmetics containing anionic surfactants in these types of products, mixtures of different surfactants, polymers, emollients, and hydrophobic substances are used (Bujak et al., 2015, 2018; Draelos et al., 2013; Fevola et al., 2010; Klimaszewska et al., 2017, 2018a, b; Seweryn, 2018; Seweryn and Bujak, 2018; Walters et al., 2012; Wasilewski et al., 2016a, b). Among natural polymers one can distinguish the proposed in the article Klimaszewska et al. (Klimaszewska et al., 2019) marine collagen with various molecular weights. According to the authors (Klimaszewska et al., 2019), it contributes to a significant reduction of the irritant potential induced by anionic surfactants. On the basis of the studies conducted on zein number, bovine albumin, and human keratinocyte cell line (Klimaszewska et al., 2019), it was found that the use of collagen in bath liquids significantly reduces the irritating effect of the composition. It was observed that with increasing molecular weight of marine collagen, the irritant effect of liquids on the skin decreases.

Marine collagen may provide an alternative to the widely known bovine and porcine collagen types. Type I of

✉ Emilia Klimaszewska
e.klimaszewska@uthrad.pl

¹ Department of Chemistry, Kazimierz Pulaski University of Technology and Humanities, 27 Bolesława Chrobrego Street, Radom, 26-600, Poland

collagen in fish is similar to that in mammals, containing three polypeptide chains, each of which consists of about 1000 amino acid residues (Addad et al., 2011; Saito et al., 2001; Sivakumar et al., 2000). Acids have a strong affinity for protein surfaces, and their moisture-binding activity contributes to the hydrating effect. Another significant aspect is that collagen of this type can be obtained from by-products of other industrial processes, *e.g.* fish skins, which are regarded as fish-processing waste (Addad et al., 2011; Saito et al., 2001; Sivakumar et al., 2000). In addition, marine collagen is characterized by high biodegradability and is sustainably sourced (Nikolovski et al., 2008; Silva et al., 2014).

It should be noted that in addition to the safety of using bath liquids for children in relation to the skin, the features associated with the functionality of these types of products are also important. Requirements for bath liquids for children are mainly related to cleaning, foaming, and rheological properties. Cleansing the skin is very important for the hygiene of the child's skin. It protects against contact with microorganisms and harmful substances (Gelmetti, 2001; Walters et al., 2008). In the case of children, the composition of cleansing cosmetics should be chosen so that it does not cause excessive lipid removal from the stratum corneum, as this may lead to increased trans epidermal water loss, lower skin hydration and even redness, dryness, and irritation or itching of the skin (Ananthapadmanabhan et al., 2004; Gelmetti, 2001; Nikolovski et al., 2008; Walters et al., 2008). These types of cosmetics should also have adequate viscosity facilitating, among others, dispensing the product from the packaging and dissolving it in water (Klimaszewska et al., 2017). In the case of infants, high foaming properties of these types of products are not desirable. Conversely, the design of cosmetics formulas for children over 3 years old should be characterized by high foaming capacity (Barel et al., 2014).

This article attempts to check the effect of various forms of marine collagen (Collagen Amino Acids—150 Da, Hydrolyzed Collagen—12,000 Da and Soluble Collagen—300,000 Da) on the stability and usable properties of these types of products. Soluble Collagen has a characteristic triple helix structure and a total content of hydroxy amino acids such as bovine collagen. It has a high level of Glycine (34.3%), Alanine (12.1%), Proline (11.9%), and Hydroxyproline (7.6%). Collagen Amino Acids contain high levels of hydrophilic amino and carboxyl groups. This results in superior moisture retention benefits, particularly at high relative humidity. However, Hydrolyzed Collagen has an analogous amino acid composition with human tissues and is the most commonly used form of collagen in the cosmetics industry (Addad et al., 2011; Klimaszewska et al., 2019; Saito et al., 2001; Sivakumar et al., 2000).

It is noteworthy that the marine collagen used in the article is derived from warm water fish (tilapia), the level of hydroxyproline (an amino acid in human skin) is much higher than that of marine collagen derived from cold water fish. Most fish collagen is contained in the skin and scales of fish. Marine collagen used in the prototypes of bath liquids for children comes from the skin of fish tilapia.

Similar to the authors' first work on the safety of using marine collagen (Klimaszewska et al., 2019), the obtained preparations were analyzed empirically, and the obtained results were correlated with the molecular mass of added collagen forms. Turbidity, dynamic viscosity, foaming properties, and the ability to emulsify fatty soils were examined.

Experimental

Materials

The raw materials used to make the prototypes of bath liquids for children are presented in Table 1:

Methods

Turbidity Test

The turbidity of bath liquids for children was assessed by the nephelometric method using the TN-100 Eutech Instruments (POL-ECO-Aparatura, Wodzisław Śląski, Poland). The measuring range includes the values of the

Table 1 Raw materials used to make the prototypes of bath liquids for children (Klimaszewska et al., 2019)

INCI name	Trade name	Company
Sodium laureth sulfate	Texapon N70	BASF, Ludwigshafen, Germany
Cocamidopropyl betaine	Dehyton K	BASF, Ludwigshafen, Germany
Collagen amino acids ($M_w = 150$ Da)	Collasurge	Croda Poland
Hydrolyzed collagen ($M_w = 12,000$ Da)	Crotein M	Croda Poland
Soluble collagen ($M_w = 300,000$ Da)	Collasol M	Croda Poland
Glycerin	Cremerglyc	Cremer Hamburg, Germany
Lactic acid	Lactic Acid	HSH Chemie, Poland
Sodium chloride	Sodium Chloride	POCH, Gliwice, Poland
Sodium benzoate and potassium sorbate	KEM BS	Pol Nil S.A., Poland
Aqua	Distilled water	

tested parameter from 0.01 to 1000 NTU (nephelometric turbidity units). The final result was an arithmetic mean of the turbidity values, from ten independent measurements.

Dynamic Viscosity

The dynamic viscosity of bath liquids for children was tested with Brook field HADV III Ultra. The measurement was carried out at the temp. of 22 °C at 10 rpm rotational speed. RV/SSRZ S Spindle SET were used. The volume of the tested samples was 250 cm³.

Foaming Properties

Foaming properties are determined according to the Polish standard PN-EN 12728: 2001. In a graduated cylinder equipped with a perforated disc mounted at the end of the rod, 100 cm³ of a 1% aqueous solution of the formulation was prepared. The foam was generated by making 60 strokes for 60 s. The foam volume was read after 10 s from the time of its rising (foaming ability). The foam stability index was calculated according to the following formula:

$$X = \frac{V_2}{V_1} \cdot 100\% \quad (1)$$

where V_1 is the volume of foam after 1 min (mL), and V_2 is the volume of foam after 10 min (mL).

Ability to Emulsify Fatty Soils

The ability to emulsify fatty soils was tested according to the Polish standard PN-C-77003: 1997. The research methodology was described by Wasilewski and co-authors (Wasilewski et al., 2016a, b).

Statistical Analysis

Each analysis of bath liquids for children was carried out in triplicate. Differences between the obtained values were analyzed using the Graph Pad Prism 5.0 software using a one-way ANOVA and the Tukey test. Assumed level of significance $\alpha = 0.05$.

Results and Discussion

Development of Formulations for Obtaining Collagen-Enriched Bath Liquids for Children

The subject of the research was the prototypes of bath liquids for children, developed on the basis of the literature

(Ananthapadmanabhan et al., 2004; Barel et al., 2014; Nikolovski et al., 2008; Walters et al., 2008, 2012) and own experiences. Concerning aspects related to the safety of these types of preparations were analyzed in Klimaszewska et al. (2017, 2018a, b, 2019). Formulation P1 is a reference liquid that does not contain marine collagen. Formulations P2–P4 differed in the form of marine collagen: Collagen Amino Acids (150 Da), Hydrolyzed Collagen (12,000 Da), and Soluble Collagen (300,000 Da) (Table 2).

The technology for obtaining bath liquids for children with marine collagen was described in work Klimaszewska et al. (2019). The pH is adjusted to 5.5 with Lactic Acid in all bath liquids.

Turbidity Test

The size of particles occurring in the solution may affect its turbidity. Turbidity is the opposite of clarity, and it is expressed in NTU. The results are shown in Fig. 1.

The turbidity value determined for the reference liquid was 5.25 NTU. Following the addition of marine collagen with a molecular mass of 12,000 Da (P3) and 150 Da (P4) to the formulation, the turbidity results were similar to those obtained for the reference bath liquid. Only the addition of marine collagen with a molecular mass of 300,000 Da produced a significant (nearly 28-fold) increase in turbidity compared to the reference liquid. However, the result obtained at the level of 150 NTU does not disqualify P4 bath liquid in terms of both stability and application. It is a slightly “opalescent” product.

The turbidity results correspond to the results of particle size determination, which were reported by Klimaszewska

Table 2 Formulations of prototypes of bath liquids for children, % by weight (Klimaszewska et al., 2019)

Component (INCI name)	Formulations of bath liquids for children			
	P1	P2	P3	P4
		Concentration (wt.%)		
Sodium laureth sulfate		6		
Cocamidopropyl betaine		2		
Glycerine		3		
Collagen amino acids		0.1		
Hydrolyzed collagen			0.1	
Soluble collagen				0.1
Sodium chloride		3		
Lactic acid		to pH \approx 5.5		
Sodium benzoate and potassium sorbate		0.5		
Aqua		to 100		

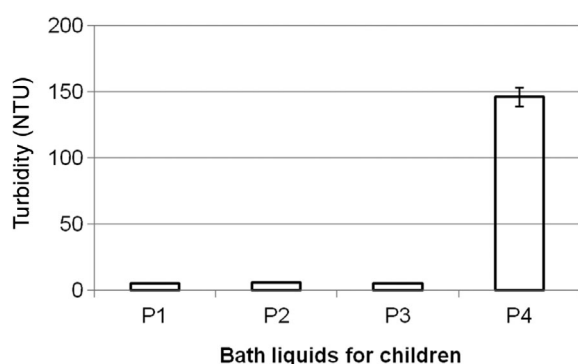


Fig. 1 Turbidity values of bath liquids for children. P1—reference liquid, without marine collagen. P2—liquid with collagen amino acids, P3—liquid with of hydrolyzed collagen, and P4—liquid with soluble collagen

et al. (2019). Larger particles of marine collagen added to the formulation P4 hinder light wave penetration through the system, which increases the turbidity of the solutions.

Dynamic Viscosity

The viscosity of bath liquids for children determines their functional properties, for example by facilitating the application of the formulation to the skin. The property is also important for ensuring the ease of dispensation from packaging. Consequently, it is a significant aspect of product quality assessment. Dynamic viscosity of bath liquids for children is shown in Fig. 2.

The lowest value of viscosity (1564 mPa s) was shown in the formulation not containing any marine collagen (P1). An addition of marine collagen to the formulations of bath liquids for children has an impact on the dynamic viscosity of the formulations under study. An increase in the molecular mass of marine collagen added to the bath liquids was found to be accompanied by an increase in dynamic

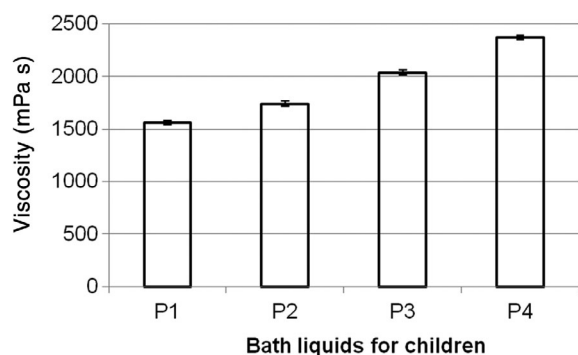


Fig. 2 Dynamic viscosity of bath liquids for children at 10 rpm rotational speed. P1—reference liquid, without marine collagen. P2—liquid with collagen amino acids, P3—liquid with of hydrolyzed collagen, and P4—liquid with soluble collagen

viscosity. The η value in the liquid P4, containing marine collagen with a molecular mass of 300,000 Da, was about 34% higher than the value noted in the reference liquid P1.

The behavior of surfactants and polymers in aqueous solutions has been widely discussed in the literature (Alves et al., 2018; Bossard et al., 2010; Deselnicu et al., 2015; Draelos et al., 2013; Gouveia et al., 2008; Koski et al., 2004; Panmai et al., 1999; Zhang et al., 2014). Findings reported by different authors show that the addition of a small amount of anionic surfactant to an aqueous solution of water-soluble polymers is followed by an increase in viscosity. For example, Alves et al. (2018) analyzed solutions of various surfactants with hydrophobically modified polyacrylic acid. At a pH level higher than 6.5, or through the binding of anionic surfactants, the polymer swells and forms a strong three-dimensional network. Also, Panmai et al. (1999) studied interactions occurring between hydrophobically modified polymers and surfactant micelles including sodium dodecyl sulfate. Interactions between hydrophobic polymers and surfactant micelles are a result of their bridging, followed by network formation, which leads to an increase in the viscosity of the solution, or even gelation. Another factor with a considerable impact on rheological properties is the molecular mass of the polymer. As the molecular mass of the polymer increases, the viscosity in aqueous solutions increases as well (Koski et al., 2004; Panmai et al., 1999), which was demonstrated, among others, by Bujak et al. (2015). The authors recorded the highest maximum viscosity values in the body wash gels with a polymer with the highest molecular mass, containing sodium chloride at 7 wt.%.

Foaming Properties

Foaming properties are among the most important functional characteristics of bath liquids intended for use in children. Although the ability to generate foam is not reflected directly in the washing performance of bath liquids, foam volume and stability are very important aspects affecting, among others, the functional characteristics of these products. The foaming properties of the bath liquids for children are shown in Fig. 3.

The foaming ability of the bath liquids for children was found to range from 510 to 570 cm³. The lowest foaming ability was observed in the reference product. In other bath liquids, containing marine collagen with different molecular mass, there was an approximately 10% increase in foaming ability compared to the reference formulation. Another parameter evaluated in the bath liquids under study was the foam stability index. The values of the parameter varied in the range of 90–96%. The highest foam stability was noted in the bath liquid P4 enriched with Soluble Collagen ($M_w = 300,000$ Da).

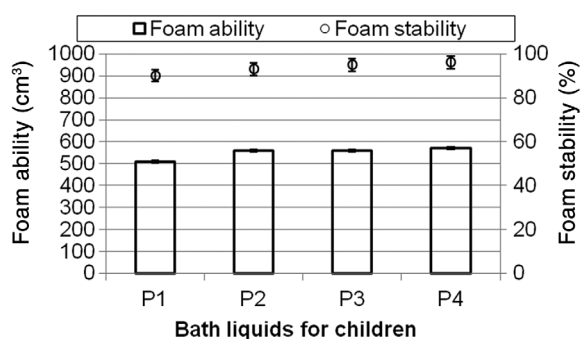


Fig. 3 Foaming properties of bath liquids for children. P1—reference liquid, without marine collagen. P2—liquid with collagen amino acids, P3—liquid with hydrolyzed collagen, and P4—liquid with soluble collagen

The effect of adding polymers to surfactant solutions has been discussed in the literature (Alves et al., 2018; Bao et al., 2008; Bujak et al., 2018; Draelos et al., 2013; Fevola et al., 2010; Koski, Yim and Shivkumar, 2004; Penfold et al., 2006; Petkova et al., 2013). Polymers, including proteins and hydrolysates, are used as foam-boosting ingredients in the food and cosmetics industries. The addition of polymers to surfactant solutions leads to the saturation of polymer chains and the formation of free micelles in the volume phase. The process increases the volume and stability of foam as a result of increased viscosity of the volume phase.

The findings of the study are consistent with the study findings reported, among others, by Bujak et al. (2015). The authors analyzed the effect of macromolecules including polyvinylpyrrolidone, polyvinylpyrrolidone/hydrolyzed wheat protein crosspolymer, and hydrolyzed wheat protein on the foaming properties of body wash gels. Higher foam volume and stability were noted in the cosmetics formulated with polymers, compared to polymer-free gels. In another study (Bujak et al., 2018), the same authors found that an increase in the molecular mass of

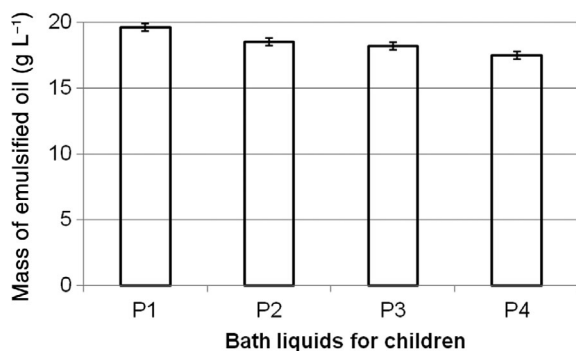


Fig. 4 Ability to emulsify fatty soils of bath liquids for children. P1—reference liquid, without marine collagen. P2—liquid with collagen amino acids, P3—liquid with hydrolyzed collagen, and P4—liquid with soluble collagen

polyvinylpyrrolidone was associated with a corresponding increase in the foaming ability of facial cleaning foam. Petkova, Tcholakova, and Denkov (Gouveia et al., 2008) studied the stability of foam in aqueous solutions of sodium dodecylsulfate and polyvinylamine, demonstrating much higher foam stability over the entire range of surfactant concentrations (0.03–0.1 mM) compared to the polymer-free solution. The authors attributed the higher stability of foam in the analyzed solutions to the formation of a dense surfactant-polymer layer on the surface of the solution, as evidenced by ellipsometry and the strongly reduced surface tension.

Ability to Emulsify Fatty Soils

Wash cosmetics are products used to maintain hygiene of the skin surface. It is connected with ensuring the appropriate level of detergent properties through the selection of its qualitative and quantitative composition. The washing mechanism takes into account a number of partial processes, in which the ability of the washing bath to emulsify fatty soils plays an important role, influencing the final effect (Miller and Raney, 1993; Rosen and Kunjappu, 2012). In the case of cosmetics dedicated for use by children, the ability to remove fatty soils should be optimal, so that on the one hand it ensures the proper washing effect and does not affect the protective layer of the lipid coat of the skin surface. The product's high ability to emulsify fats may contribute to the removal of valuable, naturally occurring hydrophobic components from the skin surface and disturb the structure of the intercellular cement of the epidermis. It may consequently lead to excessive drying of the skin and its irritation (Seweryn et al., 2018; Seweryn and Bujak, 2018).

Figure 4 shows the results of the determination of the ability to emulsify fatty soils for the tested prototypes of bath liquids for children. The form of the applied marine collagen slightly affects the ability to emulsify fatty soils. The highest mass of emulsified oil was recorded for the reference liquid—without the participation of marine collagen (19.6 g L⁻¹). As the molecular weight of marine collagen increased, a slight decrease in the ability to emulsify fatty soils was observed, by about 6–11% in relation to the reference liquid. The obtained results indicate that the raw materials of natural origin do not significantly affect the functionality of bath liquids for children in the aspect of washing effect considered in the study. On the other hand, small decreases in the ability to emulsify fatty soils may bring benefits related to the safety of these types of products in terms of their impact on the skin, in the form of the possibility to reduce the leaching of natural lipids occurring in the epidermis (Seweryn et al., 2018; Seweryn and Bujak, 2018).

Conclusion

Addition of different forms of marine collagen to the bath liquids for children based on sodium laureth sulfate was shown to affect the functional properties of these types of products. An increase in the molecular mass of marine collagen in the bath liquids produced an increase in the size of aggregates formed in water, contributing to the increased turbidity of the formulations under study. The results obtained in this work can be explained by the hypothetical mechanism proposed in the work (Klimaszewska et al., 2019), based on surfactant-polymer interactions. The highest turbidity (150 NTU) was noted for the bath liquid P4 with marine collagen ($M_w = 300,000$ Da). This value does not disqualify the P4 bath liquid in terms of both stability and use. An increase in the molecular mass of marine collagen was found to be accompanied by an increase in dynamic viscosity and foaming ability of the formulations under study. As the molecular weight of marine collagen increased, a slight decrease in the ability to emulsify fatty soils was observed. A small decrease in the ability to emulsify fatty soils may bring benefits related to the safety of these types of products in terms of their impact on the skin, in the form of the possibility to reduce the leaching of natural lipids occurring in the epidermis.

It was found that the addition of the highest molecular weight of marine collagen (300,000 Da) to children's bath liquids based on sodium laureth sulfate not only contributes to reducing the irritant effect caused by anionic surfactants, but also significantly improves the usable properties of these types of cosmetics. The proposed solution is also beneficial due to the high biodegradability of marine collagen and the possibility of replacing collagen from mammals. It should be emphasized that the production of marine collagen is ethically and ecologically balanced.

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Conflict of Interest The authors declare that they have no conflict of interest.

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Biographies

Emilia Klimaszewska is a lecturer at the Department of Chemistry, Kazimierz Pulaski University of Technology and Humanities. The area of scientific interest includes industrial research and development of innovative formulations of cosmetics, pharmaceutical products (in particular products for children), household chemistry products in the context of quality control.

Marta Ogorzałek is a lecturer at the Department of Chemistry, Kazimierz Pulaski University of Technology and Humanities. The area of scientific interest includes industrial research and development of innovative formulations of household products (especially fabric softener products) and cosmetics products, in the context of quality control.

Artur Seweryn received his M.Sc. in Chemical Engineering from the Technical University of Radom, Poland—specialization Materials Science and Engineering (2010), then Ph.D. in Commodity Science from University of Technology and Humanities in Radom, Poland in Faculty of Economics and Legal Science (2017). Currently works as a researcher in Department of Chemistry, University of Technology and Humanities in Radom. His main research interests include manufacturing of household products and cosmetics, quality assessment, aspects related to the safety of use of cosmetics and household chemicals, functional and physicochemical testing.

Tomasz Wasilewski received his M.Sc. in Chemical Engineering from the Technical University of Radom, Poland (2000), his Ph.D. in Materials Science and Engineering from the Technical University of Warsaw, Poland (2004) and his DSc. in Commodity Science from the Cracow University of Economics, Poland (2014). He was supported by the Foundation for Polish Science (2006–2007), and is currently a researcher at the Department of Chemistry, University of Technology and Humanities in Radom. Main research topics of Assoc. Prof. Tomasz Wasilewski are cosmetics and household products, their manufacturing, properties, quality assessment, and physicochemistry of aqueous solutions of surfactants. Currently, he is the Head of Department of Chemistry and the Vice Dean of the Faculty of Materials Science, Technology, and Design at University of Technology and Humanities in Radom.