USING PROCESS INSTRUMENTATION TO OBVIATE DESTRUCTIVE EXAMINATION OF CANISTERS OF HLW GLASS

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ABSTRACT

An important concern of a manufacturer of packages of solidified high-level waste (HLW) is quality assurance of the waste form. The vitrification of HLW as a borosilicate glass is considered, and, based on a reference vitrification process, it is proposed that information from process instrumentation may be used to assure quality without the need for additional information obtained by destructively examining (core drilling) canisters of glass. This follows mainly because models of product performance and process behavior must be previously established in order to confidently select the desired glass formulation, and to have confidence that the process is well enough developed to be installed and operated in a nuclear facility.

INTRODUCTION

The Department of Energy is developing waste packages that will help isolate high-level nuclear waste from the biosphere, when disposed of in a deep continental respository. In proposing regulations requiring "reasonable assurance" that the waste be isolated, the Nuclear Regulatory Commission has emphasized assurance in terms of engineered barriers. Questions about how or if isolation performance criteria should be translated to criteria pertaining to the performance of engineered barriers are not addressed here. However, "reasonable assurance" can be derived from quality assurance (QA) applied to the engineered barriers, because they can be inspected or otherwise checked for quality during manufacture or emplacement. The waste form in its processing canister is the primary engineered barrier. We can assume it will be subjected to quality assurance, not only because of its use in a repository, but because it should meet other requirements as well.

For the case of manufacture of a high level waste (HLW) borosilicate glass waste form, our objective is to describe how process instrumentation can be used to provide for quality assurance and obviate destructive examination - such as core drilling - of sealed canisters, and why this is expected to be a successful strategy. Our approach is to use basic QA concepts to trace the flow of information from performance specifications, to product specifications, and finally to process specifications, while considering at each step the control that the manufacturer actually has over the product, and to explore the reasons why this control should be adequate. We begin by discussing some basic QA concepts.

QUALITY CHARACTERISTICS

The basic objective of quality assurance is to provide evidence that the product is "fit for use."[1] In the case of waste canisters, fitness for use can be divided into several categories: handling, storage, transportation, disposal, and nuclear materials accountability. These categories allude to hypothetical individuals who would "sign off" on the acceptability of individual canisters as they move from one point to another in the waste management system. Under each category a number of "quality characteristics" must be identified, which are the attributes the product must possess to achieve fitness for use.[1] Examples of quality characteristics are:

Handling

- acceptable impact resistance, based on maximum possible free fall during handling
- acceptable gamma/neutron radiation levels, based on shielding to be provided during handling
- acceptable surface radioactivity
- acceptable size and weight, based on tolerances of handling equipment.

Storage

- acceptable thermal power
- acceptable release to water in case of canister failure, if stored in a pool
- acceptable pressurization/gas generation.

Transportation

- acceptable impact resistance, considering the protection of a shipping cask
- acceptable fire resistance, considering the cask.

Disposal

- acceptable radionuclide release behavior after repository closure, including acceptable surface area (cracking).
- acceptable radionuclide inventories
- acceptable thermal power

Nuclear Materials Accountability

- acceptable uncertainty in inventories of fissionable materials.

This list is not complete, nor is it intended to accurately reflect actual performance criteria evolving within DOE. However, it is sufficient for illustrating quality assurance approaches.

The characteristics must be subjected to quality control, which is "the process through which one measures actual quality characteristics, compares them to specifications, and acts on the difference."[1] In order to act on discrepancies with specifications, rapid measurements of quality characteristics are required. And even assuming this, the ability of the plant operator to directly control quality characteristics is required. Actually, control by the plant operator is rather limited.

QUALITY OF DESIGN AND QUALITY OF CONFORMANCE

It is important to distinguish between the two steps taken in achieving product quality. The first is to achieve "quality of design," which is the extent to which the intended product is fit for use (has the necessary quality characteristics). The second is to achieve "quality of conformance," which is the extent to which the actual product is the intended product. The plant operator has control only over quality of conformance. If the intended product is inadequate, the plant operator cannot be expected to correct it. Therefore, it is quality of conformance that will be subjected to quality control. Furthermore, many of the quality characteristics listed above are relatively insensitive to quality of conformance, compared to quality of design. For example, the canister's size, its impact resistance and fire resistance, the radiation levels and the thermal power, are all determined almost entirely by design, and actually the plant operator has little opportunity to either significantly increase or decrease the quality.

The operator's control over the plant is limited by the process variables that can be both measured and controlled. Specifically, we have considered the liquid fed ceramic melter (LFCM) process for commercial HLLW, shown schematically in Fig. 1. This is the interim reference process of DOE's Commercial Waste Treatment Program. Three tanks are shown: the HLLW holding tank, at which point the HLLW composition is determined by sampling; the feed make-up tank, at which point glass forming chemicals are added in batches and the resulting mixture is sampled; and the melter feed tank, from which the mixture is transferred into the melter at the desired rate. Both the liquid volume and the composition (from sampling) are known at the tanks, from which mass flows are inferred. The composition in the third tank must be calculated. Integration of the last two into a single tank is being considered.

A residence time period for the melter is defined by the ratio of the mass of glass in it to the mass feed rate. Typically, more than one batch of feed is mixed and transferred per melter residence time, and there is considerable convective mixing in the melter,[2] so that the composition of the glass exiting the melter is known virtually continuously. Glass is transferred to the canister by a calibrated air lift

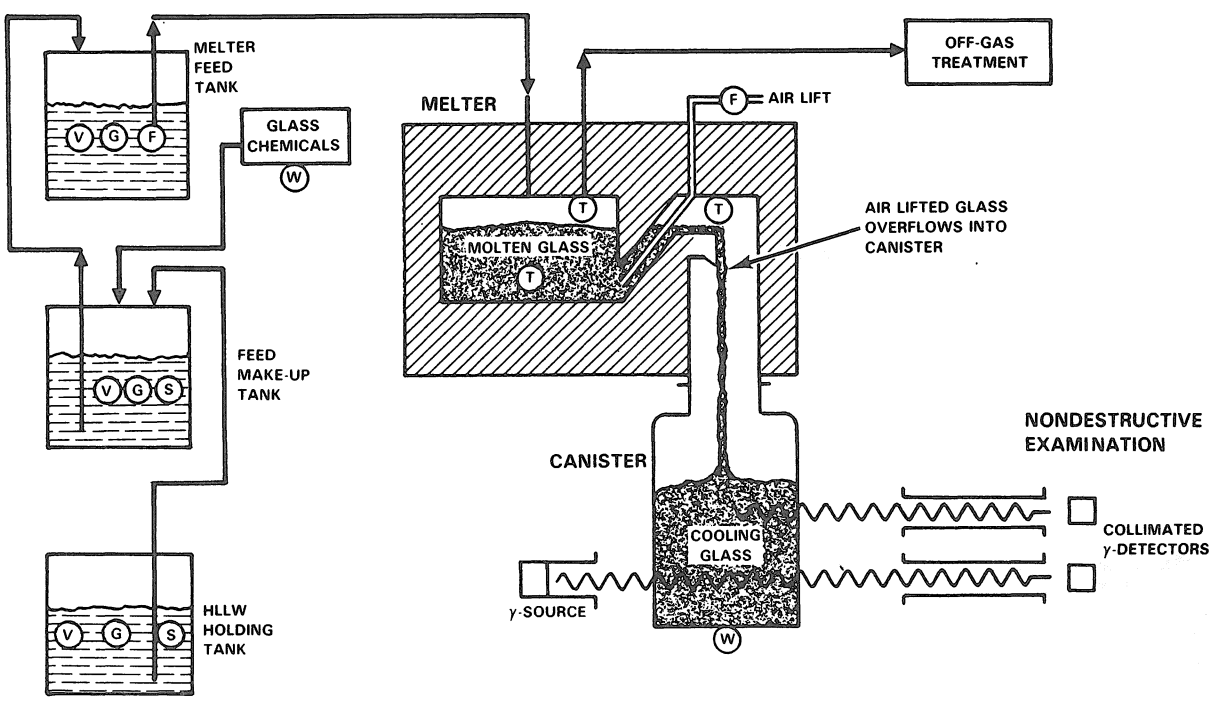


Fig. 1. Schematic of Liquid Fed Ceramic Melter (LFCM) Vitrification Process, Showing the Process Instrumentation of Interest. T= temperature, V = volume, G = specific gravity; S = liquid sampling, F = flow rate; W = weight.

(or by other means) and the temperature at the overflow point is measured. The pouring rate into a canister can also be inferred from a load cell under a canister, and by following the height of the glass using γ-scans.

By controlling flows and temperatures, the operator has some control over the composition and structure of the glass. By "structure" we mean primarily devitrification, any microcracking around devitrification crystals, and porosity, and also the "macrostructure," such as canister-scale cracking and voids. Primarily, control is over the waste loading of the glass and over the extent of devitrification and canister-scale cracking, which depend on the glass cooling rate in the canister. Devitrification and cracking cannot be controlled independently, since devitrification decreases with increasing canister cooling rate,[3] while cracking increases with cooling rate.[3,4] It is clear that the plant operator can address "quality of conformance" only in terms of the composition and structure of the glass. Therefore, a bridge must be provided between composition and structure and the quality characteristics discussed above. This bridge is a "product model," which is a set of quantitative correlations relating variations in quality characteristics to deviations in composition and structure from the intended glass.

Finally, a bridge must be provided between the controllable process variables - flow, temperature, etc. - and the composition and structure of the product. This bridge is a "process model" comprising essentially a materials balance for the feed/melter/off-gas system and a heat transfer model for glass simultaneously filling a canister and cooling to its final form. Because the process model provides, from the process measurements, a real-time determination of conformance to the intended product, it can be applied to quality control. Note that other than for the heat transfer model, neither the product model nor the process model need to be mathematically complicated.

There are two other sources of information regarding conformance to the intended product: nondestructive examination, and destructive examination (sampling). Taking samples of glass from cooled canisters can be done by coring through a canister wall and into the glass, and estimating the radial locations of the samples that are obtained. Inherently, information from sampling is not available soon enough to act on any difference between the actual and intended product, canister-by-canister.

On the other hand, nondestructive examination (NDE) can include scans of the glass in a canister as it fills, by using collimated detection of the γ-radiation emitted by the glass, and of γ-radiation from a ^{60}Co source that is transmitted through and attenuated by the glass. Combined, these serve to estimate the homogeneity of the density and the homogeneity and concentration of γ-emitters in the glass. Hence, information about composition and structure is made available during processing.

It would also be possible to fill small containers of glass mounted near the top and inside of a canister, that could be removed prior to closing the canister. This would provide samples of the glass composition, but would not be indicative of any devitrification, cracking, or voids in the actual canister. The samples woud be valuable for tracking process control. For our purposes, this can be considered delayed NDE.

USES OF PROCESS AND PRODUCT MODELS

We can now list the different kinds of elements through which we can implement process control, quality assurance, and the development of specifications. They are:

1. Sources of information prior to processing
   a. A specification of "fitness for use."
   b. A derivation of quality characteristics.
   c. A product model that correlates quality characteristics with the controllable product attributes: composition and structure.
   d. A process model that correlates composition and structure to the controllable process variables: flow rates and temperatures.
2. Specifications derived prior to processing
   a. Product specification - derived from quality characteristics using the product model.
   b. Process specification - derived from the product specification using the process model.
3. Sources of information during processing
   a. Process measurements.
   b. NDE (γ-scans).
4. Action taken during processing
   a. Operation of the process.
   b. Comparison of process data with process specifications.
   c. Comparison of product data with product specifications.
5. Sources of information after processing
   a. NDE (e.g., weld leak check, calorimetry, delayed composition samples).
   b. Destructive examination (core drilling).

These elements are assembled in each of Fig. 2 through 5. The figures show how the elements are used to provide specifications, process control, quality control, and quality assurance. Each involves a different flow of information among the same elements. Figure 2 portrays the flow of information among elements that occurs before processing: quality characteristics are derived from a specified fitness for use, then product specifications are derived from them using the product model, and then process specifications are derived using the process model.

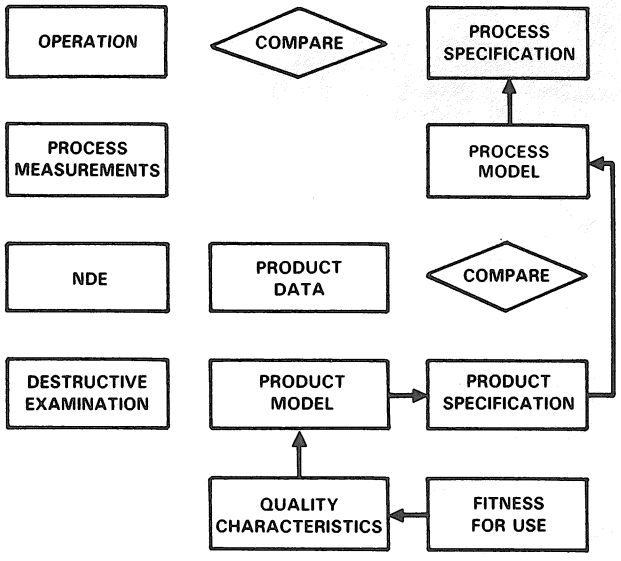


Fig. 2. Diagram of flow of information that establishes product and process specifications before processing.

Figure 3 portrays the flow of information that implements process control: process data is compared to process specifications, and any difference is acted on by changing the operation of the process. This results in new process data, and the usual process control loop obtains. This loop will presumably be automated.

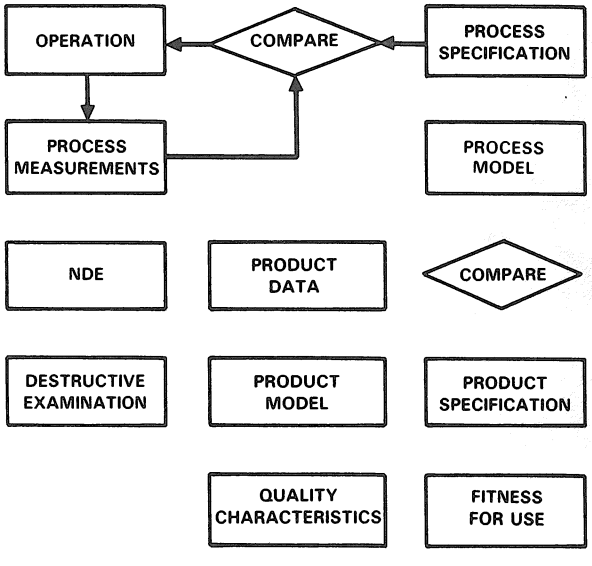


Fig. 3. Diagram of flow of information that constitutes process control during processing.

Figure 4 portrays the flow of information that implements quality control, in the context of that which can be controlled by the plant operator: quality of conformance. Product data is inferred primarily through the process model. Since NDE can immediately add to the product data, then a quality control loop occurs which includes process control as its major, but not sole, constituent. With respect to NDE, the loop proceeds from product data to a comparison with product specifications, and thence any difference is acted on by using the process model to adjust the process specifications. This then influences the operation of the process, and the loop is closed. Therefore, to the extent that NDE adds to product data beyond that already inferred using the process model, the process model is tested during processing.

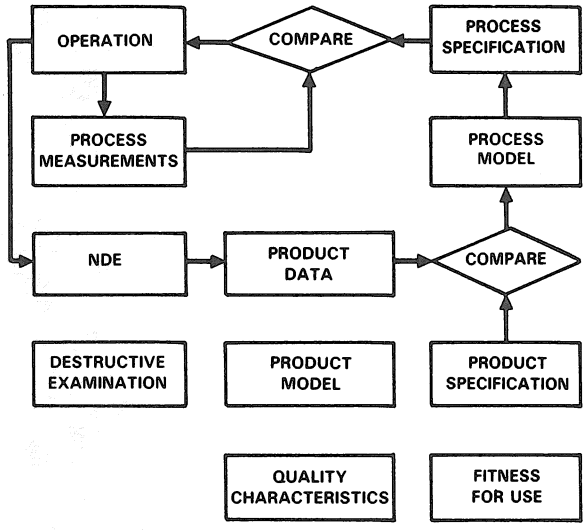


Fig. 4. Diagram of flow of information that achieves quality control during processing.

The above applies to the assumed objective of quality control, canister-by-canister. Quality control lot-by-lot would consist of examining one lot of canisters for either composition and structure or directly for quality characteristics, and then making a decision on changing process specifications, or even product specifications, for the next lot. Some of the tests that could be conceived, such as actual drop tests, could be so destructive as to require a canister to be "recycled," although it is not known how this would be done. Nominally, canisters that are cored after being welded shut are no longer fit for use, but of course could be resealed by welding metal plugs in place. However, voids would be introduced, and it would be speculative as to how much cracking, for example, was caused by coring the canister.

We regard destructive examination of canisters as the approach of last resort for quality control, since:

1. It is undesirable to produce any significant number of canisters without confidence that each canister can be sent to a repository.

2. The sampling and analysis would require hot cells for coring and repairs, additional shielded facilities and equipment to analyze the samples, and an attendant increase in cost and maintenance.

Figure 5 portrays the flow of information that implements quality assurance. Product data are inferred from process measurements using the process model. Additional information about the

product is available from both NDE and, if desired, from destructive examination. Quality characteristics are then inferred from product data using the product model. Similarly, additional information about quality characteristics is available from both NDE and, if desired, from destructive examination. In particular, NDE can be used to measure thermal power (calorimetry), radiation field, dimensions, surface contamination, and weld integrity. Also, the waste loading and homogeneity of waste loading can be estimated from γ-scans, which supplement mass balances in determining the composition.

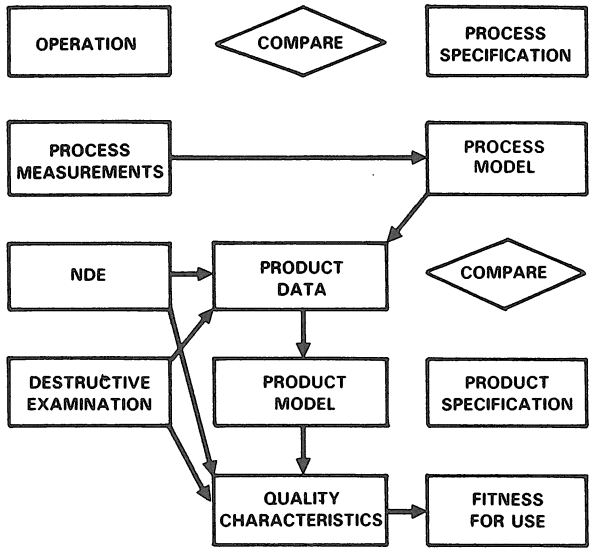


Fig. 5. Diagram of flow of information that achieves quality assurance after processing. It is proposed that the contribution of "destructive examination" is unnecessary.

OBVIATING DESTRUCTIVE EXAMINATION

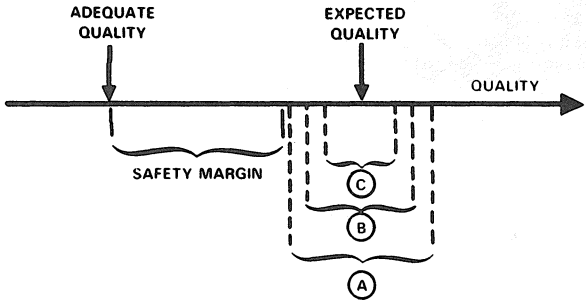
Of the three sources of information available to achieve quality assurance - process information, NDE, and destructive examination - we feel that destructive examination should not be instituted, for several reasons. First, it is important to recognize that sampling cannot reduce to zero the uncertainty in either the glass's quality characteristics or its composition and structure. If enough samples are taken, the (nonzero) variance throughout a product can be estimated from the variance among the samples. Of course, it is desirable to minimize the amount of sampling. If few enough samples per canister are taken, independent information (viz., the process model) is needed to estimate how representative the samples are of the remaining glass in the canister.

Second, some quality characteristics cannot be determined by sampling. Some involve the entire canister, such as impact resistance. Others, such as cracking, are influenced by the sampling itself, particularly as would be manifest in the samples.

Third, measurements of quality characteristics that would be feasible to perform on a multitude of radioactive samples may not be sophisticated enough to credibly contradict alternative predictions of quality. For example, if the product's composition and structure are known, then an alternative prediction of quality is available using the product model. This comment applies specifically to the "leach resistance" of the glass. The predicted release of radionuclides from a glass in a repository environment over thousands of years is an important aspect of "fitness for use" to be determined in the course of adopting a particular glass formulation, which then becomes the intended product. This determination follows from a program of testing and mathematical modeling, after which acceptability is conferred on the composition and structure of the glass that is reproduced throughout the program. It seems infeasible to equivalently determine acceptability for each of the many radioactive samples that would arise if sampling were relied on for quality assurance of the waste form. Therefore, demonstrating that one has produced the intended glass - i.e., the same composition and structure as was reproduced throughout the testing program that was the basis for adopting the glass - becomes the objective, in this case.

Finally, it seems unreasonable to assume that destructive examination would be necessary to reduce the uncertainty in the product quality to an acceptable level. This follows because of an inherent need to establish adequate process control and process specifications, simply so that the product is rarely out of conformance with product specifications. In order for the plant operator to be assured that an adequate process has been engineered, the process model and product model must have been established well enough to both derive specifications and to predict the product quality with an acceptably small uncertainty. Figure 6 illustrates conceptually the relationships among the expected quality, the limit of acceptable quality, and the uncertainty in quality as determined first solely by process measurements, and then by adding NDE, and finally by further adding destructive examination, as sources of information. The scale is arbitrary and for illustrative purposes only. We have depicted the uncertainty based only on process measurements as being small compared to the difference between expected and required quality, resulting in a margin of safety. This means that it is not necessary to apply sampling toward further decreasing the uncertainty. If this were not true, e.g., if the largest uncertainty overlapped the limit of acceptable quality, then the operator would be obviously risking nonconformance when the quality of the product was ultimately revealed, since only process measurements (and limited NDE) would be available to control the process, and hence to control the quality of the product.

In a perhaps more typical industrial setting where lots can be tested and either recycled or thrown away, and where specifications are often strict, destructive examination can be both feasible and necessary. However, in the case of QA applied to canisters of HLW glass, it would be difficult to literally recycle a canister, and they cannot be thrown away. Also, some of the most important quality characteristics, such as radionuclide release rate, derive from regulations applicable to the thousands of canisters in a repository, collectively. Then statistically, the required variance per canister will be much less stringent. Many of the



(A) UNCERTAINTY BASED ONLY ON PROCESS MEASUREMENTS

(B) LESSER UNCERTAINTY IF NDE IS ADDED TO (A)

(C) MINIMUM UNCERTAINTY IF DESTRUCTIVE EXAMINATION IS ADDED TO (B)

Fig. 6. Expected relationship between uncertainty and quality.

other quality characteristics will be much less sensitive to quality of conformance than to quality of design, as is discussed above.

Aside from considerations of quality, there is also a need to verify simply that the process "works"; i.e., that it can be operated for extended periods without upsets. For example, operations can be disrupted by foaming[5] or precipitation[6] in the melter, if sufficiently adverse combinations of composition and temperature are allowed to occur there. Therefore, process control must be developed, characterized, and demonstrated for this reason alone.

Given the above, we can expect that a process model and product model sufficient to satisfy process control requirements will also be sufficient to achieve the relationship between quality and uncertainty that is illustrated in Fig. 6. Furthermore, obviating the need for destructive examination by using these models will place no burden on those who characterize glass quality or who engineer the vitrification process, beyond that imposed by the prior need to verify that the process is understood well enough to proceed with installation.

CONCLUSIONS

1. Product quality divides into quality of design (the extent to which the intended product is acceptable) and quality of conformance (the extent to which the actual product is the intended product). A vitrification plant operator has control over quality of conformance. Quality of design must be previously established in choosing a glass formulation and a canister design.

2. Product specifications must be written in terms of attributes that can be controlled by the plant operator. These are the composition and structure (devitrification, cracking) of the glass.

3. There are few direct measurements of product quality that can be made during processing. Therefore, quality control of individual canisters must be accomplished primarily through process control. The links between process control and quality control are a process model that predicts the achieved composition and structure from the controllable process variables, and a product model that predicts product quality (i.e., product performance) from its composition and structure.

4. Because the process model and product model will have been previously developed to assure that the process is adequately controlled, they will also be available to adequately infer product quality from process measurements and from nondestructive examination, for the purpose of implementing quality assurance. Therefore, obtaining still more information about product quality by destructively examining canisters should be unnecessary. By recognizing this and carefully documenting the demonstration of process and product models, one should be able to rely on process instrumentation to obviate destructive examination of canisters and the attending increase in facilities, maintenance, and cost.

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